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# SYMPOSIUM PROCEEDINGS: ENVIRONMENTAL ASPECTS OF FUEL CONVERSION TECHNOLOGY (MAY 1974, ST. LOUIS, MISSOURI)

RESEARCH TRIANGLE INSTITUTE

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Franklin A. Ayer (Compiler)

Research Triangle Institute
P.O. Box 12194
Research Triangle Park, North Carolina 27709

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Control Systems Laboratory
National Environmental Research Center
Research Triangle Park, North Carolina 27711

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October 1974

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# FOREWORD

The proceedings for the symposium on "Environmental Aspects of Fuel Conversion Technology," is the final report submitted to the Control Systems Laboratory for the Environmental Protection Agency Contract No. 68-02-1325. The symposium was held at the Chase-Park Plaza Hotel, St. Louis, Missouri, on 13-15 May 1974.

The principal objective of this symposium was to review and discuss environmentally related information of coal conversion technology. More specifically, papers were presented that covered environmental quality and standards, fuel contaminants, environmental aspects of specific fuel conversion systems, fuel utilization and total environmental assessment, and research and development needs.

Mr. T. Kelly Janes, Chief, Fuel Processing Section, Clean Fuels and Energy Branch, Control Systems Laboratory, National Environmental Research Center, Environmental Protection Agency, Research Triangle Park, North Carolina, was the General Chairman of the Symposium.

Mr. William J. Rhodes, Fuel Processing Section, Clean Fuels and Energy Branch, Control Systems Laboratory, National Environmental Research Center, Environmental Protection Agency, Research Triangle Park, North Carolina, was the Project Officer and Technical Coordinator of the Symposium.

Mr. Franklin A. Ayer, Manager, Environmental Technology Department, Center for Technology Applications, Research Triangle Institute, Research Triangle Park, North Carolina, was the Symposium Coordinator and Compiler of the proceedings.

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13 May 1974

## Opening Session

T. Kelly Janes
General Chairman

## (KEYNOTE ADDRESS)

# ENVIRONMENTAL ASPECTS OF FUEL CONVERSION TECHNOLOGY

**John K. Burchard***

Good morning, ladies and gentlemen. It is my sincere pleasure to welcome you to our first symposium on the environmental aspects of fuel conversion technology. I do not believe that it is necessary, especially with this group, to justify or explain the importance to our Nation of establishing a viable industry for the conversion of coal to gas and liquid fuel. The Federal Government has recently identified massive financial support to accelerate the development of this technology. American industry and universities are also undertaking a broad research program to meet this challenge.

However, of equal importance to the overall public health and welfare is the need to insure that these new energy technologies are accomplished in an environmentally sound manner. The development of this or any technology that results in additional damage to the environment will necessarily label us as remiss in our responsibilities and as lacking in technical skill. The Environmental Protection Agency has the primary responsibility in the Federal Government for assessment of the associated environmental problems and for aiding their solution through the appropriate pollution control techniques; however, it is quite apparent from the magnitude and complexity of the technology and problems involved, that the environmental protection requirements will demand a sincere cooperative effort by *all* those engaged in this undertaking. The continued investigation of pollutants causing adverse health effects and ecological damage is mandatory in order to provide the basis for assessing the impact of process emissions. This information is needed for the identification of the necessary control techniques and the development of suitable control methods, so that the process developers can meet their environmental requirements in a timely and cost-effective manner.

Meetings such as this should enable those engaged in the development of conversion process technology, and those engaged in the development of environmental safeguards, to exchange ideas and goals, and to identify the needed research and cooperation. Only with such cooperation can our respective objectives be attained without a lamentable waste of time and money.

The Control Systems Laboratory (CSL) has had, for some time now, a vital interest in the emission of pollutants from the use of coal. Our efforts have included work in flue gas desulfurization, combustion phenomena, physical and chemical coal cleaning, and coal conversion technology. In 1969, we initiated a study of advanced power cycle technologies to define how the Nation's increasing demand for electrical energy could be met with maximum efficiencies and minimum insult to the environment. This study reviewed the technical attributes as well as disadvantages of topping and bottoming cycles, closed- and open-cycle gas turbines, and combined gas and steam turbine systems. The combined gas and steam turbine cycle showed potential as a very attractive technique and led us into the area of low Btu gasification and associated pollution problems. In 1970, the need for high temperature and pressure fuel gas cleanup systems was identified, and work was initiated on control techniques for sulfur-based compounds. In 1972, CSL initiated a broad study to assess the environmental impact of coal conversion technology. This will be greatly expanded in the near future.

As with any vital organization, our program has grown and changed course over the years in response to changing needs and priorities. Currently, our activities include the general pursuit of control techniques for:

sulfur oxides,
nitrogen oxides,
particulate material, and
hazardous and toxic air pollutants.

The part of our program specifically concerned with fossil-fuel conversion processes involves support of:

Fluidized-bed combustion;
Characterization of fossil fuels by types and levels of pollutants;
Environmental assessment of such areas as coal

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*Director, Control Systems Laboratory, Environmental Protection Agency, Research Triangle Park, North Carolina.

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gasification and liquefaction, advanced power cycles, and shale oil;

Physical and chemical desulfurization of coal; and

General combustion research studies.

This is a broad program, and it has given us an opportunity to gain both the process and environmental perspectives. We initiated this symposium because our studies have clearly illustrated the need for increased information exchange and cooperative efforts. We plan to sponsor such meetings periodically as a forum for the many organizations involved in this field.

Fuel conversion technology is quite complex even by itself; the additional technology that will be required for adequate pollution control adds significantly to this complexity and could have a major effect on cost. EPA is concerned about both factors. Our goal is to aid in achieving the most environmentally sound systems at the lowest possible overall cost. It should be readily apparent that by the time these plants become a commercial reality, the pollution control required will be more comprehensive and stringent than that now in existence.

Although sulfur compounds are of major concern, the emissions of various other trace pollutants will also have to be controlled. The thermal treatment of coal inevitably results in the formation of numerous organic compounds, many of which are potentially carcinogenic. For example, several types of polynuclear aromatics have been identified in emissions from coke ovens. Trace metals such as vanadium, cadmium, and mercury are also worrisome. The list of potential pollutants can be almost endless. When one considers the forecasts of future coal consumption for conversion processes, the possible total emissions into the environment become very significant.

The problem becomes even more obvious when it is realized that the main energy utilization systems for the foreseeable future are to be developed on a high priority basis within the next 5 to 10 years by a massive Government-financed R&D program. Within this time frame, it will be humanly impossible to quantify adequately all of the pollutants of concern and their associated health effects. Thus it will be impossible to precisely define the pollutant control levels required; the best safeguard is to have an active pollution control program which will be developing the necessary new and improved control methods concurrently with the development of the new energy systems. If we wait until the energy technologies are already in place, it will be extremely difficult and expensive to retrofit them with improved pollution control equipment, and our country will suffer the consequences accordingly.

This symposium is divided into four major areas:

The rationale for environmental standards,

Known and potential problems,

The status of present developments, and

Needed research and development.

During these sessions, the informational, as well as technological, gap areas should become evident. The challenge is here and will rapidly become more visible. It is up to all of us to accept this challenge, to meet it, and to solve it.

In conclusion, I again welcome each of you and hope that these next few days will be interesting and productive.

13 May 1974

Session I:

## ENVIRONMENTAL QUALITY AND STANDARDS

William J. Rhodes
Session Chairman

# ENVIRONMENTAL QUALITY AND STANDARDS FOR AIR

Jack R. Farmer*

*Abstract*

*The key provisions of the Clean Air Act of 1970 are discussed as they affect fuel conversion processes. Primary emphasis is placed on Section 111, standards of performance for new stationary sources, and the role these standards play in the national strategy for air quality management. The current standards, the standards development process, and plans for future standards are also discussed.*

The Environmental Protection Agency's authority to control the discharge of pollutants into the atmosphere is provided in the Clean Air Act, as amended in 1970. As far as stationary sources are concerned, the act contains several regulatory and enforcement options which can be used to control air pollutants. These options include (1) national ambient air quality standards/State implementation plans, (2) standards of performance for new stationary sources, and (3) national emission standards for hazardous air pollutants.

The national ambient air quality standard/State implementation plan provisions are contained in sections 109 and 110 of the act. These provisions require the Administrator to set national primary and secondary ambient air quality standards and require the States to adopt and submit plans for achieving such standards. Primary standards indicate those levels of air quality which are necessary to protect public health. Secondary standards indicate those levels which are necessary to protect public welfare. National standards have been issued for particulate matter, sulfur dioxide, nitrogen oxides, oxidants, hydrocarbons, and carbon monoxide. After the Administrator issues a national ambient air quality standard, States have 9 months to develop and submit an implementation plan which includes emission limitations on existing sources which will provide for the attainment of the national standards. The Administrator must review and approve or disapprove the State plans and promulgate substitute provisions for disapproved plans. This review can result in a combination of State and Federal regulatory action. The plans must provide for the attainment of primary standards within 3 years and secondary standards within a reasonable time. The Administrator may approve a 2-year extention for achieving primary standards if necessary technology is not available.

On May 31, 1972, the Administrator approved or disapproved all of the State implementation plans. At that time, EPA recognized that it would not be possible to meet all of the sulfur oxides control requirements in these plans within the time frame required by the act. Because of physical limitations on our ability to clean the emissions of high-sulfur fuels on a large scale in the time permitted by the act, achievement of the particulars of the State plans would require the availability of large additional supplies of "clean" fuels—natural gas and low-sulfur coal and oil. Unfortunately, our long-overdue concern for air quality comes at a time when the abundance of clean fuels in the United States is rapidly disappearing and energy experts are becoming worried about our ability to meet our energy needs, even independent of environmental considerations.

Our per capita energy consumption is the highest in the world—twice as much as that for Great Britain; 2½ times as much as that for Germany; 4½ times as much as that for Japan. Our average annual energy demand has risen from 2.2 percent for the 1955-1960 period to 5.1 percent for the 1965-1970 period—only 10 years later. In 1972, petroleum demand alone grew by 7.2 percent. The significance of this is starkly outlined when we reflect on the fact that a 4-percent growth rate doubles demand every 17 years, and a 7-percent growth rate doubles demand every 10 years.

In the fall of 1972, EPA determined that the implementation plans would jeopardize the production of more than 100 million tons of U.S. coal. By delaying or modifying regulations more stringent than those needed to meet primary ambient standards, this coal could be utilized. Accordingly, for the past year EPA has been working with the States to delay imposing those regulations which are designed to protect against property damage and are stricter than those necessary for attaining health standards. This policy permits more coal to be burned and frees clean coal and scarce stack gas cleaning equipment to be used in areas where they are needed to meet the more important health standards.

Obviously, developing processes which will convert high-sulfur or "dirty" fuels into low-sulfur or "clean"

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fuels will help in attaining the national ambient standards. As the shortage of clean fuels like natural gas worsens, there will be an increased demand for more plants to produce clean fuels from dirty fuels.

Section 111 of the act directs the Administrator to issue emission standards for new or modified sources which may contribute significantly to air pollution causing or contributing to the endangerment of public health or welfare. The emission standards must reflect the best degree of control (taking cost into consideration) which EPA feels has been adequately demonstrated. As an overriding consideration, no new plant, even if it meets the new source performance standards, can be built if it will cause a violation of a national ambient standard. To date, new source performance standards have been issued for 12 source categories. These include fossil-fuel-fired steam generators, portland cement plants, incinerators, nitric acid plants, sulfuric acid plants, asphalt concrete plants, petroleum refineries, storage vessels for petroleum liquids, secondary lead smelters, secondary brass and bronze ingot production plants, iron and steel plants, and sewage sludge incinerators.

Section 111 also authorizes the Administrator to issue standards of performance for sources of pollutants which have not been covered by a national ambient standard or a hazardous air pollutant standard. These pollutants are referred to as "designated pollutants." This standard of performance requires the States to submit a plan to EPA with enforceable emission regulations applicable to existing sources of the type covered by the standard of performance. An example of this is sulfuric acid mist from sulfuric acid plants. The national ambient standard covers sulfur dioxide but not sulfuric acid mist. States have not yet submitted their plans for sulfuric acid mist from sulfuric acid plants because EPA has not issued regulations or guidelines specifying the requirements for such plans. Draft regulations have been prepared and are currently being reviewed within the Agency. These regulations will be ready for proposal in the *Federal Register* in the very near future.

The final important section of the act which deals with stationary sources is section 112, which directs the Administrator to issue national emission standards for hazardous air pollutants. A hazardous air pollutant will cause an increase in mortality or an increase in the serious irreversible, or incapacitating reversible, illness. These pollutants are considered to be a more serious threat to public health than pollutants covered by national ambient standards. The hazardous pollutant emission standards must provide an ample margin of safety to protect public health. The act does not require the consideration of cost or availability of control technology in determining the allowable emissions for hazardous pollutants. These emission standards are effective upon proposal for new sources and within 90 days after promulgation for existing sources. Waivers of compliance may be granted to existing sources for up to 2 years. Hazardous pollutant standards were issued for asbestos, beryllium, and mercury on April 6, 1973.

It seems obvious from examining the provisions of the act that Congress established a dichotomous approach whereby emissions from existing stationary sources are controlled to an acceptable level by the State implementation plans and the emissions from new stationary sources are to be stringently controlled by new source performance standards to prevent new air pollution problems.

EPA has developed a national strategy for air quality management utilizing the implementation plans and new source performance standards provisions of the act. The strategy is designed to result in an optimum regulatory effort. Hazardous pollutants are considered a special case and will be handled whenever data become available that indicate a specific pollutant is hazardous.

As indicated earlier, new source performance standards have been issued for only 12 source categories. An accelerated program for new source performance standards is being planned which will result in more standards in a shorter time period. There are several objectives which the new source performance standards program will achieve. Emphasis will be placed on the following areas: (1) nitrogen oxides, (2) hydrocarbons (oxidants), (3) sources reviewed in accordance with significant deterioration regulations, (4) emerging industrial processes, (5) augmentation of the air quality management approach, and (6) designated pollutants.

(1) *Nitrogen Oxides.*

Currently, stationary and mobile sources each contribute about 50 percent of the Nation's nitrogen oxides emissions. The mobile source control program recently proposed to Congress will cause a reduction in total nitrogen oxides emissions from automobiles for the next 20 years, despite growth. There is no corresponding mandatory program for stationary sources, and ambient nitrogen dioxide standards cannot be maintained in most cities with only automobile control. By 1985, the proposed automobile standards will reduce nitrogen oxides emissions so that stationary sources will contribute

70 to 80 percent of the nitrogen oxides emissions. This and other information indicate the need and cost effectiveness of an intensive control program for stationary sources of nitrogen oxides.

Since many control procedures for nitrogen oxides involve basic burner designs, they are applicable only to new sources. Such an intensive new source performance standard program for all important nitrogen oxides sources is assumed in EPA's request to Congress to delay implementation of the 1976 automobile standard of 0.4 gram per mile. To implement it will require increased research and development on control technology and a change in the Clean Air Act to allow equipment standards as well as the emission standards now allowed by section 111. Such a program is of first priority in the new source performance standards program, although standard setting and peak work loads will not yet occur for several years.

(2) *Hydrocarbons (oxidants).*

The hydrocarbon/oxidant relationship is very complex. Current understandings include:

(a) Air quality standards will be very difficult to achieve in most large cities;

(b) Nonurban oxidant concentrations far exceed the national ambient standard throughout the summer in many parts of the country;

(c) These concentrations are above natural background levels and may be related to atmospheric loading of hydrocarbons over large areas;

(d) Stationary sources currently contribute 12 million tons of hydrocarbons annually (over 40 percent of emissions nationwide), and under existing control programs this will increase to 14 million tons in 1985 (over 70 percent of the total).

A recent analysis by EPA shows that the national ambient standard for oxidants will not be maintained in many urban areas without increased control of stationary sources of hydrocarbons. Also, the standard in nonurban areas will not be attained solely by control of automobiles nor by transportation control plans at selected sites. In many areas, it is becoming obvious that maximum control of all stationary sources of hydrocarbons is the best way to complement the automobile standards in handling oxidant problems. A near-term objective of the new source performance standards program is to set standards for all source categories where control is achieved by conservation measures, i.e., controlling evaporation and discouraging solvent usage or substituting for it. Decisions on standards for sources requiring control by afterburners will be delayed until the extent of the need can be better determined. Such a program is analogous to the automobile control program and is a necessary complement to it.

(3) *Prevention of Significant Deterioration.*

All regulations and concepts being considered to prevent significant deterioration of air quality rely on the inclusion of applying available control technology on major new sources to minimize deterioration. New source performance standards provide a formal, publicly developed, specific definition of available technology for these large sources and will be a key factor in the implementation of regulations. Standards will be promulgated as soon as possible for all sources listed for review in whatever significant deterioration regulations are finally promulgated.

(4) *Emerging Industrial Processes.*

The emergence of new industrial processes, such as coal gasification, use of oil shale, and gas turbines for power generation, present control problems that demand early study and documentation. State agencies often do not possess the capability to reach decisions on control in such cases. Emerging industrial processes therefore present a class of source categories for which new source performance standards may play a unique role and are a continuing necessity. The early use of new source performance standards for such processes avoids future problems in terms of nonuniform State/local regulations and provides early direction to the industry.

(5) *Augmentation of the Air Quality Management Approach.*

The major regulatory thrust of the Clean Air Act is a program of air quality management. Theoretically, the benefits of air quality management are great. Under this concept, control is required only when needed and only to the extent needed. Basically, the air quality management approach is required for the attainment and maintenance of the national ambient air quality standards. For several reasons, this approach is tenuous for particulate matter and oxidants. The air quality standard for particulate matter cannot yet consider size of particles nor chemical composition; all sources are not well known (e.g., fugitive, reentrainment, aerosols formed in the atmosphere from gases); and background levels are a problem. Poor understanding of the hydrocarbon/oxidant relationship, the present inability to attain source receptor modeling, and natural background levels are current problems for oxidant strategies. Although implementation plans based on current knowledge must be pursued under the act, mastery of air pollution caused by particulate matter and oxidants may be a longer term affair and may eventually depend simply on widespread application of good source control. In this regard, new source performance standards operate like automobile standards. They are the least costly ways of applying source controls over the long term. They allow considerations of economic impact at the time the standards are set, they minimize the possibility of continually changing emission limitations, and they provide information on control needs at the time that industry makes decisions on expansion. Future new source performance standards for sources of particulate matter will emphasize control of fine particulates, toxic metals, and fugitive pollutant sources.

(6) *Designated Pollutants.*

For some pollutants that are emitted from only a few types of sources or that are a problem in only a few sections of the country, the full program of setting a national ambient air quality standard and requiring States to submit implementation plans is not warranted. New source performance standards and the regulation of existing sources by States through the mechanism of section 111(d) will provide a complete national strategy for these pollutants. After analysis of the options, EPA has determined that control of fluorides is needed and that it should be handled under section 111; therefore, new source performance standards will be proposed in the very near future for most significant fluoride sources.

An EPA program for setting new source performance standards on fuel conversion processes is well underway. The present emphasis is on regulating sulfur emissions; plans are not yet firm for developing standards for hydrocarbons, nitrogen oxides, particulate matter, or hazardous pollutants. Booz, Allen, & Hamilton is under contract to identify the technology that could be applied to remove and recover sulfur compounds from gases produced in fuel conversion facilities. This investigation will also assess the emission reduction and the associated costs that will result from applying this technology. The completion of the contract is scheduled for November 1974. Battelle Columbus Laboratories is also under contract to identify and assess alternative control strategies that could be applied to regulate sulfur emissions from fuel conversion plants and to determine the potential emissions from the installation of fuel conversion facilities. This contract is scheduled to be completed in July 1974.

Plans have been made to propose new source performance standards for plants which convert coal to substitute natural gas by March 1975 and for plants that convert coal to low-Btu gas for use as an industrial fuel by September 1975. Projects are being initiated to study processes for converting oil shale and tar sands to gas and for liquefying coal; however, a schedule has not yet been developed for proposing such standards.

# ENVIRONMENTAL QUALITY AND STANDARDS FOR WATER

**Kenneth M. Mackenthun** *

*Abstract*

*The Federal Water Pollution Control Act Amendments of 1972, P.L. 92-500, provide a framework of comprehensive and interrelated laws and actions for the protection, preservation, and enhancement of the water environment. The national goal as expressed in this comprehensive legislative package is that, wherever attainable, by 1983 water quality should provide for the protection and propagation of fish, shellfish, and wildlife, and for recreation in and on the water. Furthermore, the national policy is that the discharge of toxic pollutants in toxic amounts be prohibited. Water quality criteria provide the baseline of the protection pyramid for the aquatic environment. Water quality criteria are levels of particular constituents in water that, when not exceeded, will insure successful propagation and continuation of aquatic life significant for the preservation of the integrity of the aquatic resource. From such criteria, ambient quality standards may be developed as an enforcement tool in water resource preservation. Likewise, effluent limitations or standards for toxic materials and for other pollutants are included as provisions of P.L. 92-500 to insure that the receiving waterway is protected against violation of the criteria necessary for its continued survival. In addition to a foundation of good laws, regulations, and enforcement capabilities, society needs yet another goal. There is a need for an environmental ethic that reflects the ecological conscience and conviction of individual and corporate responsibilities and that would help water, land, and air in its natural selfrenewal. The solution to environmental problems facing society today is the heart of survival itself.*

Contemporary man is beset with many complex water-associated environmental problems, each of which seems of such urgency that solutions must be found. Society has long witnessed the results of environmental neglect. We have seen environmental problems grow in magnitude, seemingly feeding upon themselves, as in uncontrolled cancer.

Man's continuing struggle to reach a viable compatibility with his environment has attained a significant and accelerating degree of concern in recent times. The term "ecology," which initially was used to denote the relationship of an organism with its environment, has now been broadened in popular usage to include nearly any and all environmentally related problems. Formerly, such words as "ecology" and "eutrophication" were used by a scientific few only; now they are commonplace dinner-table words. As citizens, we are concerned about the accelerating degradation of our environment as well as about clean water, clean air, and the lasting quality and quantity of our resource base. As a society, we are confronted with mountains of solid wastes, some of which spill into our waterways; mountains of sewage sludge that must reach an ultimate disposal site; the discharge, spill, or dumping of toxic or hazardous materials; the quality and quantity of the Nation's streams, rivers, estuaries, and wetlands; as well as the maintenance and the renewability of the vast oceanic resource, which is the ultimate receptor of many of the Nation's waste products.

Environmental controls for abating environmental insults can be obtained in an appropriate and in an adequate fashion. Such controls can be achieved through a combination of: (1) strong legislative and governmental units to propose, enact or promulgate, and enforce laws and regulations; (2) citizens' concern and consciousness, and their desire to improve the environment and leave it a better place in which to live, although by so doing, it may result in changed life styles; (3) the identification, investigation, research, and demonstration of environmental pollution and its treatment or control; and (4) the application of the best in technology to recycle, reduce in quantity, and treat to an adequate degree the wastewaters arising from municipal, industrial, agricultural, and other activities.

Generally, society has enacted laws adequate to provide controls for major pollutants. Such laws had their genesis in the early 1900's; by the 1930's and the 1940's, good pollution laws began to appear as State statutes. Federal laws, likewise, have been getting more stringent throughout time.

On January 1, 1970, the National Environmental Policy Act, Public Law 91-190, was signed into law.

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This act directed all Federal agencies to consider the environmental consequences of any proposed action and decisionmaking. Specifically, it required a detailed statement of the environmental impact or assessment of each proposed action significantly affecting the quality of the human environment. Such a statement must address any adverse environmental effects which cannot be avoided should the proposed action be implemented, alternatives to the proposed action, and any irreversible or irretrievable commitments of resources that would be involved in the proposed action. At the time of its enactment, it was hailed as one of the most significant Federal laws relating to the protection of environmental quality. That appraisal has not dimmed appreciably today.

The ocean-dumping bill was another significant piece of Federal water pollution control legislation. This law, the Marine Protection, Research, and Sanctuaries Act of 1972, Public Law 92-532, was enacted October 23, 1972. The act prohibited the dumping of any radiological, chemical- or biological-warfare agents or any high-level radioactive wastes into the ocean. It established an EPA permit system for the control and dumping of other materials into designated ocean dump sites. Permits that are issued for ocean dumping are based on criteria that take into account the effects of such dumping on fisheries resources, plankton, shellfish, wildlife, shorelines, and beaches. The act provided that the Corps of Engineers would permit the dumping of dredged spoils pursuant to criteria developed by the Environmental Protection Agency for the protection of the oceanic resource.

The Federal Water Pollution Control Act Amendments of 1972, Public Law 92-500, were enacted into law on October 18, 1972. This very comprehensive water pollution control legislation established a nationwide system of permits, aimed at controlling the discharge of pollutants from all point sources into the Nation's waters. Industrial point-source waste discharges must be provided with the best practicable control technology currently available by July 1, 1977, and the best available technology economically achievable by July 1, 1983. Domestic sewage must receive secondary treatment by July 1, 1977, with a higher degree of waste recovery at a future date. The granting of Federal funds to support States and communities in sewage treatment plant construction was accelerated by the act. Other highlights of the act addressed nonpoint source wastes, the discharge of toxic materials, water quality criteria and standards, effluent limitations, pretreatment standards for industries entering municipal sewage treatment plants, and the restoration of eutrophic lakes.

A century ago, the famous French physiologist, Claude Bernard, wrote, "True science teaches us to doubt and, in ignorance, to refrain." For many environmental issues, the crisis stage has been reached, and we have lost the alternative that would permit us to refrain from taking an action. Society must proceed steadfastly, with reasoned and considered judgment, and with wisdom to solve its environmental problems. Decisions must be based upon the state-of-the-art of contemporary knowledge. Ignorance, in the context of the quote, must be associated only with the broadest of definitions. It has been said that science at best is not wisdom; it is knowledge. Wisdom is knowledge tempered with good judgment. Good judgment is fostered by wide experience dealing with the knowledge of science and, in the case of water quality, with a broad association with water pollution problems.

The act provided a number of separate points of attack to control, preserve, and enhance water quality. Significant among these are provisions for developing water quality criteria, the institution of water quality standards, the development and enforcement of effluent restrictions for toxic materials, and effluent limitations for industrial waste categories. Quality criteria are the baseline in the pyramid of actions for the management and control of pollution within the aquatic environment. Water quality criteria are scientific facts developed from experimental or in situ observations that depict organism responses to a defined stimulus or material under identifiable or regulated environmental conditions. Such criteria are based upon the state-of-the-art in analytical capability, and in interpretive judgment at any given time. From such an assemblage of criteria, which represent experimental or in situ facts under given environmental conditions, a recommended criterion may be formulated for a specific element or material that is designed to include a sufficient degree of safety, to protect the aquatic environment, or to provide for particular uses of the water. A recommended criterion specifies a concentration level of the element or material that can be tolerated by essential aquatic life, and that will permit a continuation of life for those species.

The act required EPA to publish revised recommendations for water quality criteria by October 1973. The prime sources for these recommendations included the 1968 Report of the National Technical Advisory Committee to the Secretary of the Interior and a revised and updated version of this report to EPA prepared under contract by The National Academy of Sciences. Comments currently are being received on the proposed water quality criteria. Effort is progressing toward the development of this document into a form suitable for final publication. The goal in this endeavor is to arrive at the most adequate criteria levels under our current state of knowledge for the protection of the aquatic resource. The act states that criteria for water quality must accurately reflect the latest scientific knowledge of the kind and extent of all identifiable effects on health and welfare, including, but not limited to, plankton, fish, shellfish, wildlife, plant life, shorelines, beaches, esthetics, and recreation which may be expected from the presence of pollutants in any body of water, including groundwater. Properly prepared, water quality criteria define the limits of quality that will provide for a continuation of life in water acceptable to society and for those particular uses which man has associated with water since Biblical times.

Prior to the 1972 Amendments, the Federal Water Pollution Control Act required States to adopt, for Federal approval, and to enforce water quality standards for interstate waters. Each of these standards consisted of a designated use or uses for the water body, quality criteria, and schedules of implementation for sources discharging pollutants into the water whose achievement would result in meeting the designated criteria. The 1972 Amendments mandate an expansion of the Federal/State water quality standards system to make them applicable to all navigable waters, including intrastate waters. The act further provides that the States shall review applicable water quality standards at least once every 3 years and, as appropriate, modify and revise their content. The principal bases for the water quality standards are quality criteria.

The act provides that designated toxic materials be considered as a special category and that effluent limitations for such toxicants shall be developed that take into account the toxicity of the pollutant, its persistence, degradability, the usual or potential presence of affected organisms in receiving waters, the importance of such affected organisms, and the nature and extent of the effect of the toxic pollutant on such organisms. Again, quality criteria provided the baseline of data from which effluent limitations were developed for the toxic materials.

A discharge of heat is a pollutant under the provisions of the act and is subject to applicable effluent guidelines and to water quality standards. A special section of the act contains provisions unique to thermal discharges that (1) permit thermal control requirements to be modified if the discharger can demonstrate that such requirements are excessively stringent for the protection of aquatic life, (2) require that effluent limitations minimize the potential damage of cooling-water intake structures, and (3) provide assurances that the pollution source will not be required to make multiple modifications respecting its thermal components within a short time period.

The act provides for the definition of effluent limitations placed upon industrial categories of point-source wastes that provide the best practicable control technology currently available for 1977 implementation and the best available control technology economically feasible for 1983 implementation. In addition to the ongoing effort within the Agency to define and promulgate these technologic levels for particular industrial categories, the act provides for an Advisory Committee consisting of nine scientists and engineers qualified by education, training, and experience to provide, assess, and evaluate scientific and technical information on effluent standards and limitations. The Committee is to transmit such information to the Administrator of the EPA for his evaluation in connection with the development and promulgation of industrial effluent limitations and toxic materials standards.

Secondary treatment is a mandated uniform standard for the control of municipal sewage. Pretreatment standards for new and existing industrial sources have been written to protect the operations of the treatment works into which such industries discharge, as well as to prevent the discharge of pollutants that may be inadequately treated in the municipal treatment plant. Such pretreatment standards prohibit influents into municipal works which could cause fire, explosion, process upsets, or other operational damages.

The control of discharges into navigable waters is through the issuance of a permit whose terms will be based on effluent guidelines, toxic effluent standards for certain classes of sources, and water quality standards. The effluent guidelines provide the base-level for permit limitations. These will be revised where necessary to meet applicable toxic materials standards and water quality standards. The significant portion of the water quality standards, from the standpoint of permit issuance, is the body of water quality criteria that define quality for life in water.

The act provides for the development of criteria to control the dumping of dredge spoils into navigable waters, as well as to control the discharge of wastes into estuaries and the near-shore areas of the ocean. Provisions are made for the enforcement of standards to insure that the discharge of human body wastes from vessels receives adequate treatment. The discharge of wastes from Federal facilities is addressed in stringent language, and special provision is made for an attack on problems associated with the Nation's lakes, particularly problems of an eutrophic nature.

Laws and regulations provide a framework and a focus for national concern. They do not necessarily provide the ultimate in environmental protection. In addition to a foundation of good laws, we must develop meaningful citizens' environmental concern and consciousness in all age groups. We must develop and practice a meaningful environmental ethic, both as individuals and as a community. As Aldo Leopold wrote over a quarter of a century ago, "An ethic, ecologically, is a limitation on freedom of action in the struggle for existence..." Leopold knew of what he wrote. He was a renowned professor of wildlife management at the University of Wisconsin, and he knew the land and the plants and animals that resided thereon. He said that an environmental ethic must reflect the existence of an ecological conscience and a conviction of individual and corporate responsibilities for the capability of the water, land, and air environments for self-renewal. Such an ethic must embody ethical and esthetical qualities in addition to the essential economic and technologic considerations. The integrity, quality of life, stability, and beauty of the biotic community and the environment that supports it must be preserved. As Leopold said many years ago, "A decision is wrong when it tends not to foster these principles."

Today, as never before, society is in need of an environmental ethic on the part of the residents on this planet Earth. The solution to environmental problems that confronts us as a group is the heart of survival itself. The sages of yesterday were aware of this fact. Today's man on the street is convinced of its truth.

# COAL CONVERSION TECHNOLOGY AND SOLID WASTE DISPOSAL: TIME TO TAKE STOCK

**A. Blakeman Early, Esq.***

*Abstract*

*The growth of demand of energy is rising at an alarming rate. As coal conversion technologies are developed and applied to meet this growth, significant disposal problems caused by a diminishing number of disposal sites and greater environmental requirements must be confronted. The technologies which generate the wastes which are the least harmful and are in the smallest volumes deserve attention. When conversion technologies are applied commercially, adequate disposal for solid wastes generated many years in the future must be secured as soon as possible.*

This symposium on "Environmental Aspects of Fuel Conversion Technology" is reflective of the revolutionary change in perception which has taken place over the past several years in our Nation's views on the uses and abuses of science and technology. One might designate Earth Day 1970 as the date when, as a nation, we discovered that we must confront and solve some important questions in our headlong drive toward progress and random growth in technology, industrialization, urbanization, and population. How can we adjust our sense of priorities to insure that we fulfill our energy, transportation, housing, recreation, and personal consumer needs, without intensifying resource depletion, environmental, and public health problems that we did not anticipate and that we do not want? Some people, the young in particular, are so shocked by the mounting number of problems that they seem willing to reject the benefits of technology in order to solve the byproduct problems, which heretofore had been viewed as being outside the total structure of our complex industrial society.

Even though subsequent celebrations of Earth Day have by no means reached the peak of fervor that characterized the first one, that occasion marked the beginning of what may be called a worldwide environmental revolution.

I use the word "revolution" because the movement so seriously challenges so many powerful cultural values deriving from the original industrial revolution. In this country, it has already yielded, among other things, important new legislation at all levels of government, major changes in the attitudes, and, to a significant but lesser extent, changes in the practices and activities of government, industry, and individuals. The U.S. Environmental Protection Agency is itself a product of the new wave of environmental awareness which crested as this decade began.

I hope I have not given the impression that everything has gone smoothly and that everyone has fallen into step to march toward a new world in which we use technology and science more wisely than we have in the past, without complaint and without disruption. To the contrary, there has been a lot of complaint and considerable disruption; but on the whole, the changes in institutions and attitudes that have come about in our society since 1970, in both the private and public sectors, are a remarkable testimony to the validity of the new environmental awareness that first surfaced in that year.

The current energy shortage, which of course has repercussions throughout the world, has given great encouragement to many who apparently are unwilling to face the fact that the future, when man will have the capacity to destroy the world in one great bang or slightly more slowly through continued environmental mismanagement, is **now**.

Our per capita energy consumption is the highest in the world—twice that of Great Britian, two and a half times that of Germany, and four and a half times that of Japan. Our average annual increase in energy demand has risen from 2.2 percent level for the 1955-60 period to 5.1 percent level for the 1965-70 period—only years later. In 1972, petroleum demand alone grew by 7.2 percent. The significance of this is starkly outlined when we reflect on the fact that a 4-percent growth rate doubles demand every 17 years, and a 7-percent growth rate doubles demand every 10 years.

Even official energy consumption projections to the year 2000, based on a 3.5 percent average annual growth rate, yield the following implications for the year 2000: a trebling of energy requirements, a doubling of per capita use, and an increase to 900 nuclear reactors from the 39 now operating.

Oil imports could increase to twice our entire domestic output today. Imports would approximate 25 million barrels per day—five times the current levels—and oil spills could proportionately increase. Given such import levels, the effects on our Nation's foreign policy could be devastating. All of these conditions can be expected by the turn of this century if energy consumption grows at only two-thirds of the average annual rate of the last decade.

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These projections, based on Department of Interior figures, make it quite clear that the silver lining in the energy cloud may indeed be the unpleasant shock which will wake us to some of the important realities of our time. Thus, the importance of developing coal conversion technologies has been brought to the forefront by rising demand for energy and then reemphasized by the Arab oil embargo, which demonstrated that this Nation could not remain dependent on oil imports to meet the demand.

The importance of developing other energy-producing technologies has been likewise emphasized. The Office of Solid Waste Management Programs, in which I serve, has initiated a number of demonstration projects designed to recover significant amounts of energy from mixed municipal waste, as well as to recover important materials.

About 125 million tons (1971) of solid waste are generated each year from homes and commercial establishments (offices, stores) across the country. About 70 to 80 percent of this is combustible and can be converted into energy through modern technology.

Some of this technology is being demonstrated by EPA. EPA's demonstration project in St. Louis, Missouri, converts solid waste into a low sulfur fuel that can be used as a supplement to coal in power plant boilers. Every ton of solid waste can be converted into 900 kilowatts of electricity.

Two other EPA-supported projects, one in Baltimore and one in San Diego, will convert solid waste into a combustible gas or oil using a pyrolysis process. Operations begin in late 1974 or 1975.

If energy recovery were practiced in all SMSA's in the United States, about 800 trillion Btu's would be recovered annually. This corresponds to the energy in about 4,000,000 barrels of oil per day. By comparison, this is equal to 5½ percent of the fuel requirements of all electric utilities, 12 percent of the coal used by electric utilities, and about 1 percent of all the energy consumed in the United States in 1970.

In the area of solid waste collection and disposal, our efforts also encourage a significant savings in fuel by providing information and technical assistance to cities for the purpose of enhancing productivity and efficiency in the collection and disposal of solid waste.

The future for coal conversion technologies appears to be a bright and active one because coal is the one energy resource which is abundant in domestic reserves and because much of the coal contains too much sulfur and other pollutants to be burned directly without causing unacceptable levels of air pollution unless more efficient and less costly post-combustion cleanup processes are developed. Whether the pollutants are removed before this valuable resource is burned for its Btu value or afterwards, they must be disposed of in an environmentally sound manner.

EPA is concerned about the environmental impact from the land disposal of mounting volumes of air and water pollution control residuals. These volumes are not insignificant. Air and water pollution control residues could rise from the 28 million tons generated in 1971 to 170 million tons in 1985 (ref. 1). The hazardous wastes attributable to these residues could rise from 11,000 tons in 1971 to at least 51,000 tons in 1985, not including radioactive wastes (ref. 2). The vast majority of these residues will be generated by processes designed to control particulate and sulfur oxide emissions. Control of nitrogen oxide emissions is unlikely to create significant amounts of solid residues, even though relatively large amounts of these pollutants are discharged following combustion, since the reduction of excess nitrogen oxides duirng combustion or low-temperature combustion are the techniques most likely to be used. Neither process results in the creation of solid waste residues (ref. 3).

Most coal conversion processes are in the early stages of development such that the volume and nature of residues that will be generated cannot be predicted with much accuracy. It is assumed for purposes of this discussion that the nature and volumes of these residues will approximate those of conventional coal burning processes. Indeed, several coal conversion processes will be using conventional scrubber systems to clean flue gases as well as the product gas prior to distribution. Consequently, a look at land disposal problems associated with conventional coal burning processes is likely to provide a useful indication of potential land disposal problems to be faced by coal conversion processes.

At present, wet lime/limestone scrubbing systems constitute the great majority of full-size power plant desulfurization processes. These systems are efficient and require relatively low capital and operating costs. Also, they have a longer history of development.

Variations of the process involve the use of either lime or limestone as the reactant which is carried into contact with the flue gas in an aqueous liquor. This liquor is either totally recycled (termed a cyclic operation) or totally disposed of (termed a noncyclic operation) to a reservoir. The noncyclic operation can pose some obvious water pollution problems caused by leaching to surface and ground waters. Consequently, it is believed that the large majority of systems will use a cyclic operation. Nevertheless, the solid in the liquor must be settled out before it can be reused, and this sludge must be disposed of on the land. These sludges contain calcium hydroxide, calcium carbonate, calcium sulfate, and calcium sulfite. Where efficient particulate removal is not performed upstream of the scrubber, such sludges can contain large quantities of coal ash. These liquors, in equilibrium with sludge materials, typically have dissolved solid contents of approximately 3,000 to 15,000 ppm (ref. 4). The two major methods of disposal are dewatering the sludge and trucking it offsite for landfilling, or pumping the sludge into a pond designed for permanent retention. Sulfur residues are produced at approximately a 50 percent greater dry weight amount than the fly ash normally produced by a conventional coal burning utility. This means that the sludge and ash throwaway requirement will be about 2.5 times the normal coal ash disposal tonnage. Therefore, large storage capacities are going to be necessary for disposal of these sludges. For instance, a 1,000 MWe unit over a 20-year lifetime will require about 1.6 square miles of land for disposal, assuming a wet sludge containing 50 percent solids was ponded to a height of 10 feet (ref. 5).

It is estimated that conventional coal burning processes will generate 51 million tons of ash by 1975, 56 million tons by 1980, and possibly 72 million tons by 1985. Therefore, it is well that those developing coal conversion processes share some concern for the land disposal implications of these volumes.

These volumes will be escalating at a time when capacity for landfilling solid waste is declining nationwide. It must be noted that many coal conversion processes will be competing with conventional coal burning processes as well as with other generators of solid waste for the available landfill capacity. Last year in a survey of cities having populations greater than 10,000 conducted by the National League of Cities and the U.S. Conference of Mayors, 46.5 percent of the respondents indicated that current solid waste disposal capacity would be exhausted in from 1 to 5 years (ref. 6).

My use of the term "capacity" is intended to mean capacity at disposal sites that are both located and operated in an environmentally sound manner. Flood plains, steep grades, areas above high water tables, and areas of special environmental significance are just a few of the locations where disposal sites should not be located. The basics of sound disposal site location and operation are described by EPA in its "Proposed Guidelines for Thermal Processing and Land Disposal of Solid Wastes" published in the *Federal Register* (April 27, 1973, p. 10544). These guidelines are expected to be promulgated in final form in the near future. Although they are recommendatory with regard to the public, section 211 of the Solid Waste Disposal Act, as amended, make them mandatory for Federal agencies and facilities.

In the future, disposal sites are not only going to be difficult to find, but their location and operation will be required to be conducted with greater care. For those users of lime/limestone scrubbing systems, this can prove to be a serious problem. Although there are no enforceable Federal disposal standards at this time, and State standards vary significantly, it is clear that holding ponds for scrubbing liquors and reservoirs for the long-term disposal of sludges which have not been dewatered must be carefully engineered and located, only after careful geologic and hydrologic examination of the site, in order to minimize the potential for leaching of pollutants to ground or surface waters. Moreover, initial ponding efforts indicate that the sulfate sludges have poor steeling characteristics which make it unlikely that the disposal site can be reclaimed for subsequent use. In this case, the disposer may be required to provide for monitoring and permanent care of the site to guard against future environmental harm.

Even when the sludges are successfully dewatered (problems have been experienced with mechanical dewatering in this regard), disposal by means of a sanitary landfill does not mean that such residues can then be forgotten. Preliminary EPA research has shown that a test sanitary landfill cell, 149 feet long by 30 feet wide packed with 8 feet of mixed municipal waste with a moisture content of 27.16 percent, generated 38,647 gallons of leachate over 22 months during which time 84.1 inches of rain fell (ref. 9). Attention must be given by disposers to the containment and/or treatment of such leachate generated by sanitary landfills used for residuals disposal.

EPA's concern for adequate capacity is shared by the Congress. The multitude of bills introduced over the past year concerned with solid waste disposal indicates that Congress is not satisfied with the progress the States have made without the stimulation of Federal regulation. The existing Solid Waste Disposal Act, as amended, provides only for Federal research, demonstrations, training, and technical assistance in solid waste disposal as well as for resource recovery. I would like to review briefly some of the salient features of three bills that are currently pending before the Congress and that appear to have the most significant support.

### The Resource Conservation and Recycling Incentives Act, S. 2753, introduced by Senator Philip A. Hart on November 28, 1973

The Administrator is required to issue Federal standards for the regulation of unsafe waste management practices which may include minimum performance standards with respect to the methods, techniques, and practices of waste management as well as criteria for the location, design, construction, and operation of waste treatment and disposal sites and facilities. The Administrator shall also establish standards for State waste management programs which will require State permits for generators of hazardous waste and operators of waste treatment or disposal sites or facilities of such a nature or character as to pose a significant risk to human health or the environment. Permit holders must comply with Federal/State requirements concerning information recording and reporting, transportation, storage, treatment, and transportation of wastes. If a State fails to submit an adequate program, fails to properly make such revisions, or fails to enforce its program as the Administrator may require, the Administrator shall issue regulations establishing an interim program for such States.

Standards shall be established only after considering all factors relevant to the protection of human health and the environment, including: contamination of ground or surface waters, potential hazards to transportation modes or facilities, risks associated with the decomposition of waste, and risks of surface and subsurface fires at disposal sites.

### The Comprehensive Waste Management and Resource Recovery Act, H.R. 13176, introduced by Congressman Paul G. Rogers on February 28, 1974

States must submit plans for the establishment and operation of a waste management which must include a permit system for operating a waste management facility or disposal site, the issuance and removal of which is conditioned upon compliance with standards for the location, design, construction, operation, maintenance, and abandonment of such facility or site. States must also establish a system for bringing existing and abandoned open dumps into compliance with such standards. As is stated in S. 2753, if the State fails to submit, revise, or enforce such a plan, the Administrator shall promulgate such regulations as he deems appropriate.

The Administrator shall promulgate Federal standards of performance for new sources of certain waste generation categories that may contribute significantly to the Nation's waste management problems and the endangerment of the public health or the environment. Such standards shall reflect the degree of limitation achievable through the application of the best system of reducing or eliminating the amount of toxicity of any wastes generated that has been adequately demonstrated. A new source is defined as any facility that generates wastes, the construction or modification of which is commenced after the publication of regulations prescribing a standard of performance. The Administrator may delegate the primary responsibility for enforcing such standards to any State which submits an acceptable plan for implementing them.

The Administrator would be required to establish Federal standards and procedures for the storage, treatment, and disposal of hazardous wastes which would be enforceable by means of a Federal permit system. The operation of the permit system may be delegated to any State which meets such minimum requirements that the Administrator may establish.

### The Energy and Resource Recovery Act of 1973, S. 3271, introduced by Senator Pete V. Dominici, March 29, 1973

The Administrator would be required to promulgate Federal standards for the collection, handling, disposal, and recovery of all hazardous and other solid waste which may, if improperly disposed of, cause air or water pollution or other environmental damage. Each State may, but is not required to, submit a plan for enforcing these standards within such State for approval by the Administrator. Upon approval, such States will be authorized to enforce such standards within the State.

Clearly, a pattern emerges from the foregoing description. At least some members of Congress are convinced that EPA should be empowered to issue standards to insure that solid waste is disposed of in an environmentally sound manner. These standards would be adopted, with necessary modifications, and enforced by the States. Although it is not possible to predict what requirements EPA might consider promulgating beyond those found in the Proposed Guidelines, suffice it to say that such requirements will serve to reduce the amount of land available for use as disposal sites.

Even though Federal legislation will not be forthcoming until some time in the future, the States have not been idle in addressing solid waste disposal problems. Although the comprehensiveness and effectiveness of some State solid waste laws and regulations vary widely, 45 States have such laws, and 29 States have some form of permit system for solid waste disposal sites. Some of these regulations are vague, but they take a significant meaning when interpreted on a case-by-case basis by State authorities. For instance, rule 314(e) of the Illinois Solid Waste Regulations dealing with landfills merely requires "adequate measures to monitor and control leachate." Commonwealth Edison has found, to its chagrin, that such "measures" can be interpreted to be fairly rigorous.

In December 1971, Commonwealth Edison retrofitted one of its cyclone boilers, which generates 177 MW, with a limestone wet scrubber system; it is located in Will County, Illinois. The scrubber produces approximately 19 tons of sludge per hour when operating at full load capacity. Even though Commonwealth Edison has spent nearly $2 million to find a sludge treatment method which will satisfy State authorities and which will allow the sludge to be disposed of offsite, it has experienced great difficulty in locating a site which would be acceptable to the State. The sludge is comprised of approximately 30 percent solids, 50 percent of which is calcium sulfite. The sulfites must be chemically stabilized or they will reabsorb moisture after being dried. State authorities feel that any site which has a permeable underlying soil or rock condition would be subject to sulfite ground seepage. The company has installed a thickening process which utilizes a 65-foot diameter thickener rather than utilizing the existing sludge pond. After processing, the sludge generated has a 40-percent solids content. Recently, the company has been experimenting with vacuum filtration and centrifugation as an additional method of concentrating the sludge to a 55-percent solids content. The sludge will then be solidified through a chemical fixation process, thus involving the addition of varying amounts of lime and fly ash to the sludge (ref. 10). Commonwealth Edison has located an abandoned quarry as a possible disposal site which would have to be lined with an impermeable material. It has an estimated capacity of about 1,000,000 cubic yards. However, at the current rate of residue production, the sludge and fly ash alone from the four boilers will fill this site in less than 5½ years, not including the disposal of the scrubber sludge. Based on this evidence, the company convincingly claims that wet scrubber waste will put intolerable pressure on the availability of acceptable disposal sites. In addition, Commonwealth Edison claims that annual sludge treatment costs will approximate $1,335,000 or $4.91/ton, while hauling and disposal costs will approximate $3.36/ton. This would add about 2 mills per kilowatt hour to the current cost of power from this unit, or 13 percent (ref. II).

Other processes are being developed which are considered to have the potential to desulfurize flue or product gases on a full-scale commercial basis, and to have relatively less significant disposal problems. Indeed, the three processes of magnesium oxide scrubbing, catalytic oxidation, and wet sodium-base scrubbing have the potential for producing large quantities of sulfuric acid which could be sold commercially, if market conditions were ripe. Wet sodium-base scrubbing may also lend itself to the application of technology to reduce the $SO_2$ to elemental sulfur which could either be sold or disposed of with fewer concerns for potential environmental degradation. In this regard, the coal conversion technologies that produce elemental sulfur will have a distinct advantage over other processes (ref. 12).

The foregoing discussion illustrates some of the potential problems coal conversion technologies may face concerning waste disposal on land. It may be that by the time these processes are in operation commercially, many of the disposal problems will be solved. It does seem advisable, however, that those involved in the development of these processes should provide for the wastes which will be produced and should further develop and utilize technologies which will generate these wastes in a form that will enable their disposal on the land in the least volume and in the safest manner possible. Those people involved in the commercial application of coal conversion technologies are advised to devote attention to obtaining disposal capacity well in advance of need.

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# ENVIRONMENTAL IMPACT STATEMENT REQUIREMENTS AS RELATED TO FUEL CONVERSION TECHNOLOGIES

**Sheldon Meyers***

Good morning. I cannot tell you how delighted I was when I learned that a governmental organization was going to sponsor a symposium on the environmental aspects of fuel conversion technology. It is, of course, quite proper that the Government organization that is sponsoring the symposium is the Control Systems Laboratory of the Environmental Protection Agency.

What with the recent crisis in the supply of energy, I was somewhat concerned that all the stops would be pulled out in favor of unrestrained development of new energy sources. When one is waiting in line for gasoline for an hour or so, I do not think that there is any particular sense of urgency related to protecting the environment. But then again, crisis situations have never been conducive to rational decisionmaking.

The question of interest resolves itself quite simply as to whether or not those officials, public or private, who are responsible for making decisions respecting new fuel conversion technologies should factor environmental considerations into the decisionmaking formula.

From my perspective, there can be no responsible position other than that which is in favor of considering environmental values. But let me make it absolutely clear, I am not advocating that decisions respecting fuel conversion technologies be made after considering *only* environmental factors, but that the environment be considered along with the traditional factors, such as technology and economics, before such decisions are made.

It is particularly important to assess the environmental impact of major programs such as coal gasification and oil shale production in the aggregate, as well as on a plant-by-plant basis.

I mentioned earlier the decisions to be made by both public and private officials. Let me speak first of the simpler case--the public sector.

## *Public Sector*

The public sector is simpler, of course, because of the National Environmental Policy Act of 1969--a law which is binding on all Federal agencies who take major actions significantly affecting the environment, requiring them to prepare environmental impact statements.

## *The National Environmental Policy Act of 1969 (NEPA)*

Let me briefly discuss the genesis of the environmental impact statement (EIS). It is a creature of section 102(2)(c) of the National Environmental Policy Act of 1969 (NEPA). This section states, in part, that all agencies of the Federal Government shall:

> Include in every recommendation or report on proposals for legislation and other major Federal actions significantly affecting the quality of the human environment, a detailed statement by the responsible official on:
>
> 1. The environmental impact of the proposed action;
> 2. Any adverse environmental effects which cannot be avoided, should the proposal be implemented;
> 3. Alternatives to the proposed action;
> 4. The relationship between local short-term uses of man's environment and the maintenance and enhancement of long-term productivity; and
> 5. Any irreversible and irretrievable commitments of resources which would be involved in the proposed action, should it be implemented.

Section 102(2)(c) also requires the responsible Federal official, prior to making the detailed statement, to consult with and to obtain the comments of any Federal agency which has jurisdiction by law or special expertise with respect to any environmental impact involved. Copies of the statements and comments thereon shall be made available to the president, to the Council on Environmental Quality (CEQ), and to the public. This section along with implementing guidelines published by CEQ in the *Federal Register* describe the content and mechanics of what we now call environmental impact statements.

Some additional points concerning the NEPA--first, the intent of the Congress in enacting the legislation. Section 101(2) of the act clearly annunciates such intent as follows:

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> The Congress, recognizing the profound impact of man's activity on the interrelations of all components of the natural environment . . . declares that it is the continuing policy of the Federal Government . . . to use all practiceable means and measures . . . to create and maintain conditions under which man and nature can exist in productive harmony, and to fulfill the social, economic, and other requirements of present and future generations of Americans.

Second, the intent of the Administration in concurring with the legislation. The President, in signing the NEPA on January 1, 1970, declared that the 70's

> absolutely must be the years when America pays its debt to the past by reclaiming the purity of its air, its water, and our living environment. It is literally now or never.

We see then, that the executive and legislative branches of Government shared a unanimity of purpose with regard to environmental protection. *What of the judicial branch?* Subsequent events have shown the courts to be vigorous proponents of the environment by virtue of their decisions related to the NEPA.

Third and most important, there is no provision in the act for any Federal agency to veto the projects of any other Federal agency. Those projects which thus far have been enjoined by the courts for failure to comply with NEPA were the result of procedural difficiencies in the EIS rather than from postulated environmental damage associated with the project.

So much for background. Has the preparation of several thousands EIS's, since the enactment of the NEPA, had a beneficial effect on the environment? Or has the preparation of EIS's been an exercise in bureaucratic paperwork serving only to needlessly delay critical projects?

## *Environmental Impact Statements*

As with many things, the effectiveness of EIS's has been variable. In many instances, preparation of the EIS for a particular project has had absolutely no effect on the project. In some cases, projects have been delayed, in others alterations were made which were environmentally beneficial, and there are examples of projects which have been cancelled.

There have been a number of court decisions, such as Calvert Cliffs, Gillham Dam, and Kalur which have drastically affected the internal operations of a number of Federal agencies. There is no doubt that the preparation of an EIS for a project which has been underway for sometime can adversely affect the schedule of completion, assuming that it is not cancelled entirely. There are Federal agencies involved in such projects--the completion of which will have a critical impact on both the economy and our well-being.

I concede that there is a problem with projects that are in various stages of completion. However, I am confident that solutions can be developed which will accommodate these acknowledged near-term problems. It is the longer-term use of EIS's that will ultimately contribute to determining the kind of environment in which we all live. The EIS can be used in a project's formulative stages in much the same manner as is technical and economic information. Thus, it is in the planning stages of a project that the EIS will prove its ultimate usefulness as a mechanism for protecting the environment.

## *EIS and Fuel Conversion Technology*

Since the National Environmental Policy Act and its impact statement requirements are binding only on the Federal establishment, then only those programs with some sort of Federal involvement will be affected. The Federal involvement goes beyond Federal funding; it also affects:

1. use of Federal lands,
2. requirement of a Federal permit or license.

Thus, such projects as nuclear power plants, costing on the order of $500 million of private utility capital, require an EIS because the AEC must license the construction and operation of such plants. Also, the oil shale leases in Colorado and the drilling for oil in the Outer Continental Shelf, which are being developed completely with private capital, require an EIS because of the permits issued by the Department of the Interior.

A properly prepared EIS will provide a wealth of information to management that would otherwise not be available, thus giving it the opportunity to analyze a number of options and to weigh the economic benefit against environmental cost for each of these options, before making a decision. In the past, decisions were made on major projects which turned out to be environmental disasters, in part, due to the lack of pertinent information. It has been my experience that, given the necessary information, responsible Federal managers will decide in favor of the least environmentally damaging option even though it may be a higher cost option.

### *Private Sector*

What of the private sector, where the manager has no legally binding mandate to factor the environmental amenities into the decisionmaking process? Let us also assume that there is no Federal involvement, to wit:

1. The program involves only private lands,
2. No Federal permits or license required,
3. No Federal funds involved,
4. The program is exclusively within the boundaries of a particular State and does not cross Federal lands.

Under these circumstances, no EIS will be required. But does that mean that a comprehensive environmental analysis should not be done? I submit to you that the answer should be *no*--that is to say that the responsible private official should not make his decision on technical and economic considerations only but should also analyze the environmental effects of the proposed project and its alternatives prior to making his decision. I am, of course, proposing an analysis similar to that required of the Federal establishment pursuant to the NEPA. If after completing the environmental analysis, the private sector manager decides that the program should proceed, even in the face of environmental damage; then, at least, it will have been a conscious decision. Such analyses, for example, are done by the world bank prior to making major loans, even though they are not obliged to do so by law.

How then should those of you involved in new fuel conversion technologies view the added factor of environmental analysis in the decisionmaking process? Is it something of benefit deserving the application of valuable monetary and personnel resources, or, on the other hand, is it an obstacle that must be overcome or avoided completely? I trust that an intelligent review of all the pertinent issues will bring you to the same conclusion which I have reached; that is, the environmental analysis is clearly desirable and moreover in the national interest, particularly if we feel any responsibility at all for the future well-being of the citizens of this country.

13 May 1974

## Session II:

## FUEL CONTAMINANTS

René R. Bertrand
Session Chairman

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# PROBLEMS IN THE CHEMISTRY AND STRUCTURE OF COALS AS RELATED TO POLLUTANTS FROM CONVERSION PROCESSES

**P. H. Given***

*Abstract*

*In the present state of knowledge, one can only make predictions of the probable pollution problems that will arise in coal conversion processes. The predictions must be based on (a) details of individual conversion processes, including ancillary operations, (b) knowledge of the organic and inorganic constituents of coals, and (c) geochemistry of the different major coal-producing areas of the country. The principal types of minerals in coals are clays, quartz, carbonates, pyrite, and sometimes feldspars. Potentially polluting trace or minor elements may be associated with any of these or with the organic matter; the fate of these elements in conversion processes will depend on the nature of the association of each element as well as on its chemical properties, but available data on trace elements in coals do not include information on the type of association. Existing knowledge of the chemistry, geochemistry, petrography, and mineralogy of coals will be reviewed in relation to conversion technology in such a way as to facilitate predictions of potential pollution problems.*

In all coal conversion processes, the sulfur content of the product is much less than that of the coal. Therefore, sulfur emission is not a problem when the product fuel is burnt. It is difficult to see how any gaseous fuel produced from coal could contain significant quantities of any environmentally hazardous material. Liquid fuels might contain small quantities of undesirable trace elements; whether or not this is likely to be generally true has not been determined yet.

The matter of most concern is whether or not the conversion processes themselves are likely to give rise to toxic emissions of sulfur or trace elements. The answer to this question obviously depends partly on the nature of the process, and it will have to be determined by establishing mass balances between the input of toxic elements in the coal feed and output in the various product and byproduct streams. It will also have to be determined whether toxic elements in any solid wastes are easily leached out by waters or are tightly locked into the chemical structures. The other important factors governing the potential hazard of conversion processes are the characterisitics of the coal used as feed stock. It is with this factor that this paper is chiefly concerned.

Coals are derived from partly decayed plant material that accumulates in a peat swamp. The material includes woody parts of trees (stem, branches, roots), leaves, spores and pollen (often only the waxy outer covering of these three), some charcoal-like substance from wood charred in forest fires, and some ill-defined amorphous material. These various types of substances are still recognizable after coalification. Grains of sand and clay minerals are transported in by ground waters, and mixed with the organic material; also, pyrite may be formed *in situ* as a result of microbiological processes, particularly when the water saturating the peat is saline.

All peats, soils, river waters, etc., contain some very ill-defined substances known as humic acids. These contain a variety of oxygen-containing functional groups (carboxylic acid -COOH, phenolic -OH, carbonyl C=O), which can fix cations by ion exchange with the acid groups but can also very strongly complex a variety of cations by chelation with adjacent functional groups in the structure. Of course, the plants growing in a peat swamp, like all other organisms, need a variety of trace elements for health growth; the peat must have a mechanism for concentrating or trapping the elements needed and for making them available to the plants and micro-organisms growing in the peat. Humic acids, or parts of their structures, survive in chemically recognizable forms in lignites and subbituminous coals.

Although components of peats have a large potentiality for trapping many elements, the actual concentrations are highly variable and in fact quite low in some parts of a swamp. Suppose, for example, that the peat swamp is in a basin surrounded by hills. Rocks in the hills are eroded by natural processes, and the trace elements released (together with chemically altered mineral grains) are washed by rain and streams down into the basin. Trace elements will tend to be trapped in the margins of the swamp, and the center will then obtain only low concentrations.

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Table 1. Approximate values of some coal properties in different rank ranges

|  | Lignite | Subbituminous | High vol. bituminous |  |  | Medium Volatile | Low Volatile | Anthracite |
|---|---|---|---|---|---|---|---|---|
|  |  |  | C | B | A |  |  |  |
| % C (min. matter free) | 65-72 | 72-76 | 76-78 | 78-80 | 80-87 | 89 | 90 | 93 |
| % O | 30 | 18 | 13 | 10 | 10-4 | 3-4 | 3 | 2 |
| % O as COOH | 13-10 | 5-2 | 0 | 0 | 0 | 0 | 0 | 0 |
| % O as OH | 15-10 | 12-10 | 9 | ? | 7-3 | 1-2 | 0-1 | 0 |
| Aromatic C atoms % of total C | 50 | 65 | ? | ? | 75 | 80-85 | 85-90 | 90-95 |
| Avg. no. benzene rings/ layer | 1-2 | ? |  | 2-3 |  |  | 5? | >25? |
| Volatile matter, % | 40-50 | 35-50 | 35-45 | ? | 31-40 | 31-20 | 20-10 | <10 |
| Reflectance, % (vitrinite) | 0.2-0.3 | 0.3-0.4 | 0.5 | 0.6 | 0.6-1.0 | 1.4 | 1.8 | 4 |
| Density |  |  |  | increases |  |  |  |  |

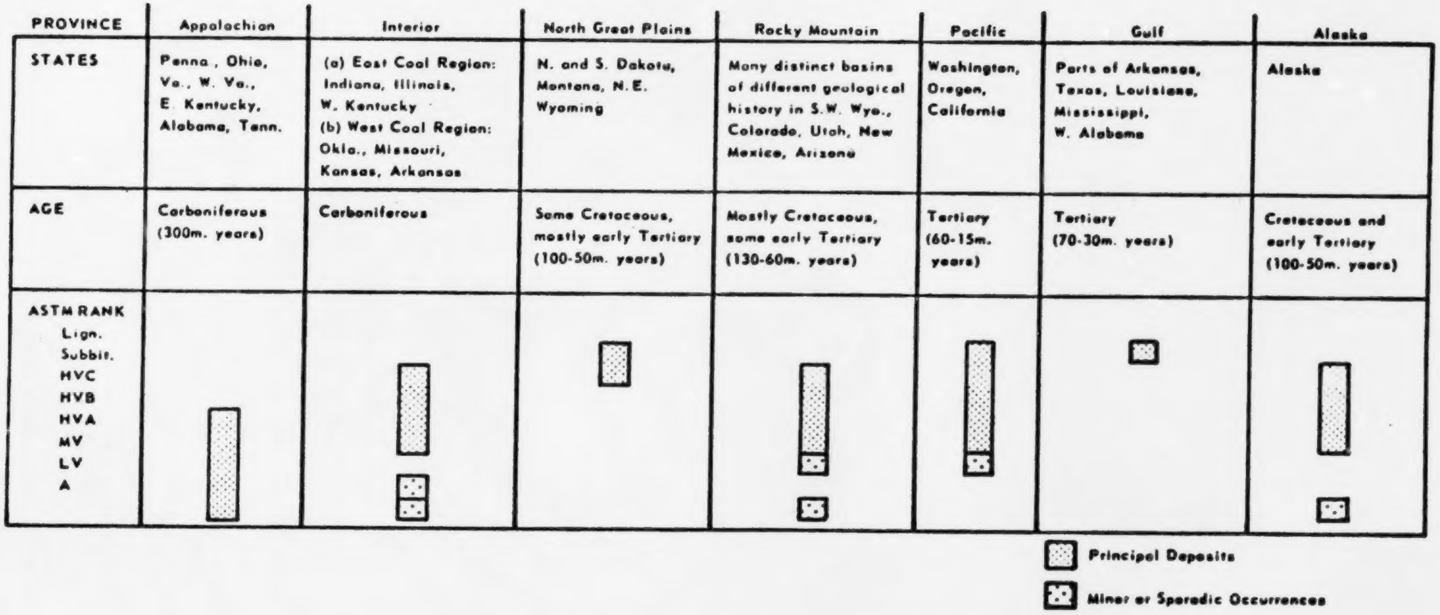
If coal formation is going to occur, sooner or later the peat gets buried under a load of other material as the area subsides. Geological processes may permit it to be buried as deep as 7,000 metres (20,000 feet), though this may take tens of millions of years to happen. At this depth, the temperature may be 200°C and the pressure may be 1,500 kg/cm$^2$. If burial is less deep, temperature and pressure will of course be lower. It is a geological fact that when deep subsidence occurs in an area, sooner or later the process is reversed, and sediments come back nearer the surface (and mountains arise in what was once a low-lying basin). The *time* of exposure of a peat stratum to elevated temperatures and pressures is therefore limited.

In practice, in many of the major coal basins, the right conditions for forming a peat swamp have recurred many times (20-50?) during the whole subsidence episode. Therefore, we may end up with a large number of superimposed coal seams, separated by varying thicknesses or inorganic rocks. The deeper the seam, the higher is the temperature-pressure regime it has experienced, and the longer the time it has been exposed to these conditions. It is exposure to elevated temperature (chiefly) for some millions of years that brings about the conversion of plant debris (peat) into coal. The more severe is the temperature-time history, the greater the degree of alteration of the organic material. In the situation considered above, where there are many superimposed coal seams, there will be a progressive change in a variety of chemical, physical, and technological properties as we examine sequentially the series of seams from top to bottom (or vice versa).

It is the extent of alteration of the organic material, associated with a progressive shift in properties, that is described by the term "rank"; high rank coals have been extensively altered. Rank is, therefore, a more-or-less continuous variable. For industrial users of coal, a continuous variable is too complicated, and classification systems are used in which the whole spectrum is divided into about 10 ranges. A coal is classified by being assigned to one of these ranges. Knowledge of the rank of a coal on the A.S.T.M. system (strictly speaking, the rank range; each A.S.T.M. class covers a range of rank) provides guidance chiefly as to (a) the ability (or lack of it) to form metallurical coke, and (b) the heating value.

Table 1 summarizes very approximately the values of a number of important chemical and physical properties for the major A.S.T.M. rank classes of coals. No figures are given for carbonyl oxygen because no reliable method for measuring it has been established, and rigorous proof even of qualitative identification is lacking. However, it is probable that some is present, and the decrease with increasing rank is rather slow, so that is may be the principal functional group in anthracites. At any rate, it is the various oxygen functional groups that can hold cations by ion exchange or chelation, and their total concentration decreases markedly with increasing rank. Therefore, organically held cations must be progressively released from a coal as its degree of metamorphism (i.e., the extent of geological alteration) increases; if released in this way, the cation may be incorporated into a mineral, but in many cases will be removed altogether in solution in ground waters.

It will also be noticed that the aromatic character of coal structure, in two distinct senses, increases with metamorphism. Much of the nonaromatic carbon appears to be present in hydroaromatic rings. It should be noted that the contents of sulfur, nitrogen, and mineral matter, and the nature of the mineral matter, do *not* vary in any systematic manner with rank. The form of combination of organic sulfur and nitrogen is not known, but it is presumed to be



| PROVINCE | Appalachian | Interior | North Great Plains | Rocky Mountain | Pacific | Gulf | Alaska |
|---|---|---|---|---|---|---|---|
| STATES | Penna., Ohio, Va., W. Va., E. Kentucky, Alabama, Tenn. | (a) East Coal Region: Indiana, Illinois, W. Kentucky<br>(b) West Coal Region: Okla., Missouri, Kansas, Arkansas | N. and S. Dakota, Montana, N.E. Wyoming | Many distinct basins of different geological history in S.W. Wyo., Colorado, Utah, New Mexico, Arizona | Washington, Oregon, California | Parts of Arkansas, Texas, Louisiana, Mississippi, W. Alabama | Alaska |
| AGE | Carboniferous (300m. years) | Carboniferous | Some Cretaceous, mostly early Tertiary (100-50m. years) | Mostly Cretaceous, some early Tertiary (130-60m. years) | Tertiary (60-15m. years) | Tertiary (70-30m. years) | Cretaceous and early Tertiary (100-50m. years) |

ASTM RANK
Lign.
Subbit.
HVC
HVB
HVA
MV
LV
A
Principal Deposits
Minor or Sporadic Occurrences

Figure 1. Distribution of coals in the United States.

largely or entirely in heterocyclic aromatic rings. The significance of reflectance will be dealt with later.

Coking behavior cannot be tolerated in some gasification processes, particularly in fluidized bed reactors. These processes must, therefore, be restricted to low rank coals, or bituminous coals must be pretreated to prevent coking; the only practicable pretreatment available at present is mild oxidation, an undesirable expedient because of the unavoidable loss of reactivity and calorific value associated with it. Moreover, part of the sulfur is released as $SO_2$.

The thermal decomposition of coals sets in at 350-400°C, and a complex mixture of aromatic hydrocarbons and heterocycles, phenols, water, ammonia and hydrogen sulfide is released at these and at higher temperatures. This condenses on cooling to two liquid phases, an organic and an aqueous phase. Since phenols are partly soluble in water, the aqueous phase contains some of these substances. The phenomena occur to some extent in coal gasification; with some processes, emission of phenol-containing waters is something that needs control. The oxidative pretreatment of bituminous coals to prevent caking is likely to have the same problem.

The coals of the United States occur in a number of distinct regions or provinces, each province having a different geological history. For this reason, it is useful to consider the coals of a province as a group, rather than to base the discussion on a geologically arbitrary division by State. Figure 1 summarizes the principal facts about these provinces, including the range of rank found in each. All of the coals of the Interior province and many of those in the Appalachian province were laid down in saline water conditions, while those in the other provinces were formed in fresh water conditions. The significance of this is that saline waters contain relatively high concentrations of sulfate ion, which is reduced by bacterial action to $H_2S$, with the result that much sulfur is fixed in both pyritic and organic forms in coals formed under marine influence. In addition, the trace elements trapped in the coals will reflect, in some sense, the complex distribution present in ocean water. The large reserves of coals in the North Great Plains and Rocky Mountain provinces contain relatively less sulfur, often less than 1 percent. It is worth noting that when the total sulfur content is above about 1 percent, roughly equal amounts of sulfur are present in pyritic and organic forms in a majority of coals. On the other hand, in coals of low total sulfur content, most of the sulfur (70-95 percent) is present in organic form.

Quantitatively, the most important minerals in the majority of coals are clays (kaolinite, illite, chlorite, montmorillonite, mixed layer montmorillonite-illites and sometimes muscovite). Feldspars are present in some western coals. In addition quartz, pyrite, and carbonate minerals (calcite dolomite, siderite, and ankerite) are common. Some secondary mineralization may occur in cleats, veins, and cracks after the peat has been buried and coal formation started; thus, some pyrite, carbonate minerals, and possibly some quartz may be emplaced after burial. Apart from this, the minerals originally deposited with the peat suffer little alteration during coalification.

As yet there is little to show, in terms of comprehensive analyses, as to whether the suites of minerals found in the coals of the various provinces are characteristic of or individual for each province. However, one can infer some answer to the question from the geological circumstances. Thus, the low rank coals of the North Great Plains province have never been deeply buried, and the over-burden on top of the seams is often an unconsolidated sediment of clays and sand. It is impossible to avoid mining some of the roof material along with the coal, and the unconsolidated sediment may yield a slow-settling mud during attempts to remove it in a preparation plant, particularly if, as is often the case, it contains mixed layer clay minerals. Some of this material may form part of the input to conversion processes.

The coals of the Rocky Mountain Province were laid down as the mountains were arising. Much of the rock forming the mountains is granite, and so the trace elements and chemically altered minerals released by the erosion of granite constituted the main inorganic input to the coals forming in basins among the foothills of the mountain range. Moreover, since the coals formed in a large number of relatively small basins, the distribution of minerals and trace elements in each is likely to be relatively uniform.

In the Appalachian and Interior provinces, the coal basins were flood plains of very large area. There were mountains along the margin of the basin, and others some way to the north of it, but they were not of the massive and granitic character of the Rockies. Some of the mineral grains were transported in water for considerable distances, and so coarser particles would have dropped out on the way. Therefore, the mineral input to the peat swamps would have had a different character from the input to the Rocky Mountain coals, and it would be more variable in different parts of the basins.

A number of publications in the literature (refs.

1-18) give information on trace elements and minerals in coals. Valuable though the data are, they provide little insight into several important issues.

Much more study needs to be made of the relation of the distribution of inorganic constituents in coals to the geology of the basins in which the coals occur, and to the geochemical principles involved. Empirical correlations of the concentrations of some trace elements with others need to be made, and, once found, a geochemical explanation may emerge. Thus, the author has noted that the concentrations of zinc and lead tend to be relatively high in areas where high pyrite contents are common; the solubility products of the sulfides of these metals are particularly low. At any rate, any generalizations that can be made about the distribution of trace elements and minerals in a seam or in the coals of a basin will reduce the problems of assessing potential environmental hazards.

Alpern and Morel (ref. 6) studied the lateral variation of trace element distributions in a number of coal seams in France, in the hope of using the distributions as fingerprints in identifying the stratigraphy of seams. However, they were forced to conclude that this was not a valid approach for the seams studied, because the lateral variations were quite large. This is not unexpected in view of the remarks made above about the trapping of trace elements in peat swamps. It conveys the warning that single values for the concentrations of trace elements in coal seams may be far from representative.

When the opening of a new mine is being considered, it is common practice in the coke-making industry to take a series of drill cores on a grid pattern in order to assess the coking properties of the coal and their lateral variation. This localizes that part of the areal extent of the seam that should be mined. It will be prudent to make similar surveys when selecting sites for coal conversion plants, and then the opportunity could be taken to assess the lateral variation of key trace element concentrations. In some seams at least, the variations may be less than indicated by the results of Alpern and Morel.

Another aspect of the distribution of trace elements in coals is the nature of their associations. Trace elements could be held in coals in one or more of the following ways (subject, in some cases, to the ionic charge and radius being suitable):

1. incorporated in the structure of clay minerals or interstitially,
2. in the clays by ion exchange with acidic OH groups,
3. by isomorphous substitution in carbonates or pyrite,
4. as a minor mineral (e.g., sphalerite, ZnS, detected in some Illinois coals),
5. as a carboxylate salt in lignites and in subbituminous coals,
6. as a chelated organometallic complex in a coal of any rank, although as already noted the extent of such complexing will decrease with increasing rank.

There is very little evidence to show the extent to which any trace element is held in each of these forms. Yet the product stream into which any trace element will go, the chemical form it will be in, and the ease with which it may be leached from the solid waste must surely depend at least partly on its association in the original coal.

These aspects of the fate of trace elements will also depend of course on the nature of the conversion process. Thus in catalytic liquefaction processes clays, quartz and carbonates will be little changed at the temperatures used (about 400°C), nor will their association with trace elements be much altered. It has been shown recently in the author's laboratory that pyrite is reduced under liquefaction conditions to the nonstoichiometric mineral pyrrhotite, $Fe_{1-x}S$, where $x = 0\text{-}0.2$. Other sulfides present, if any, might be reduced to the metal (e.g., Pb and Zn), but there is no evidence. Elements held by the organic matter in carboxylate or chelated form will be released, possibly in highly reactive form.

It is already known that titanium, which is a minor rather than a trace element in many coals, tends to accumulate on the surfaces of liquefaction catalysts and to deactivate them. Other elements, including boron, may accumulate in a similar way, and it seems likely, for kinetic reasons, that they were originally present in soluble organometallic form rather than in mineral form. Some potentially toxic elements could be scavenged in this way and tightly held on spent catalyst.

Gasification, whether to low or high Btu gas, is carried out at much higher temperatures (900-1,000°C), and profound alterations of all mineral species in coals are to be expected. All carbonates, clay minerals, and pyrite will decompose and the products of decomposition will react with each other in producing new crystal phases; this will be particularly so in the few processes in which slagging occurs. Where oxidative pretreatment is practiced with bituminous coals, sulfides will be converted to oxides, or possibly to sulfates. Organically held elements will be released in various forms. Several elements will be partially or completely vaporized (e.g., Se, Te, F, Hg, Ga, Ge). In these complex circumstances, predictions of the fate of trace elements cannot be made, and careful experimental study is needed. The information on the behavior of coal minerals in combustion processes, assembled in the excellent review by Watt (ref. 1), should provide some useful guidance to the behavior of inorganic material in gasification processes.

It was noted at the beginning of this paper that debris of the various organs of trees can be distinguished not only in peats but also after coalification. Furthermore, various processes in peats generate new materials that do not occur as such in living plants, and these materials also may be distinguishable in coals. The various kinds of bodies found in coals are termed "macerals". The most abundant maceral in most coals is vitrinite, which represents woody material that has been somewhat altered biochemically and then coalified. It is vitrinite that is responsible for the coking behavior of bituminous coals. Also, vitrinite shows a more consistent trend of properties with rank. This is why reflectance is a valuable parameter characterizing the rank of coals; using microscopic techniques, the mean reflectance of the vitrinitic maceral in sample can be measured without interference from other macerals. To measure any other property of the vitrinite, a physical separation from other macerals would have to be carried out first. In addition, the differing reflectances of the macerals are used in making a petrographic analysis of a coal sample.

In all, ten macerals are distinguished in coals. Of these, the quantitatively most important, apart from vitrinite, are sporinite (from the waxy coating of spores and pollen), fusinite (a kind of charcoal, thought to have been generated in forest fires), and micrinite (of unknown origin, highly aromatic and inert in the coking process). The few studies made so far (refs. 17,18) indicate that different suites of minerals tend to be associated with the different macerals in any one coal. The various macerals contain different concentrations of functional groups (refs. 20-22), and these may well have different spatial distributions in the organic structures. The ability to chelate trace elements or otherwise to form oranometallic complexes will therefore be different for the different macerals.

The various organic and mineral phases in coals have different hardnesses and specific gravities. In the process known as coal preparation, coals are crushed, sized, and subjected to float-and-sink separations. By no means all coal is subjected to preparation before use; when it is subjected to preparation, the objective is primarily to reduce the pyrite content, though the contents of other minerals are reduced at the same time. How effective the process is will depend upon, among other things, the particle size distribution of the minerals and the mineral-maceral associations.

Provided the characteristics of the coal are well understood, the preparation process could be so planned as to maximize the efficiency of mineral matter removal. But in addition, some degree of management of feedstock composition can be achieved with regard to the organic components also. Since these differ in their susceptibility to conversion processes, sophisticated use of preparation procedures may well be a desirable prelude to feeding coals to conversion plants.

The important point for present purposes is that any preparation of the coal before use will reduce the concentrations of some or all of the trace elements in the feedstock. There is a possibility, which should be checked by experiment, that some of the more toxic elements (e.g., Pb, Cd) are present as sulfides and closely associated with pyrite. Pyrite is the mineral of highest specific gravity in coals, so that it tends to be removed preferentially in preparation processes. On the hypothesis above, some toxic elements would be preferentially removed with the pyrite.

In conclusion, some general points about the utilization of the products of coal conversion processes should be made. It is perhaps not widely appreciated that the presence of mineral matter in the coal fired to a utility power station represents a considerable cost to the utility. Ash accumulates on steam and superheater tubes, leading to poorer heat transfer, to corrosion, and to periodical shutdowns for removal. Electrostatic precipitators for removal of fly ash are expensive in capital and operating costs; stack gas cleaning will also be expensive, if a satisfactory process is proved. Quite apart from environmental considerations, therefore, the utility industry has a substantial economic motive in desiring clean fuels.

For various technical reasons the industry is increasingly turning to gas turbines for intermediate and peak load generation. These *must* have extremely clean fuels to avoid damage to the turbine blades. The low energy efficiency of the production of high -Btu gas from coal argues against using it for power generation; if any gas from coal is used, it will be low-Btu gas. A liquid fuel from coal would be most desirable, because oil can be stockpiled for intermittent use, and the conversion process needs less water.

Centrifugation of the oil from catalytic liquefaction is easy and reduces the content of inorganic material to about 0.1 percent. This is adequate for using the oil as a substitute for a petroleum product or natural gas in package boilers in industry and for industrial process heating. This kind of use accounts for somewhat more fuel consumption in the United States than electricity generation. For firing in gas turbines, the oil will have to be filtered to reduce ash-forming constitutents to around 0.01 percent, and certain elements (e.g., Na, K, V) must be below 1 ppm. This is less easy, but can probably be done. If it *is* done, then there is no need to worry about toxic emissions when the fuel is burnt.

The four principal conclusions of this paper are:

1. From the environmental point of view, it is the conversion processes themselves that require consideration, rather than subsequent use of the fuels produced.
2. The conversion processes require study in some detail, to determine what is present in all of the product and byproduct streams and what are the possibilities of undesirable elements escaping when waste products are disposed of.
3. The inorganic constituents in coals need much more detailed and sophisticated study than they have received in the past.
4. The effect of coal preparation processes on the contents of toxic elements needs to be determined.

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# TRACE ELEMENTS AND POTENTIAL POLLUTANT EFFECTS IN FOSSIL FUELS*

H. J. Hall, G. M. Varga, and E. M. Magee†

*Abstract*

*Coal, petroleum, and shale oil are minerals whose impurities are commonly the same as the beds in which they are found. Relationships between impurity content and geographical location are well known for sulfur, but much less so for other elements. They are known in petroleum production only for sulfur, nitrogen, nickel, and vanadium. The characteristic salt water inclusions produced with crude oils are removed before further use, and they are not considered (like the minerals in coal) as a part of the product.*

*The amounts of trace elements found in coal are, for the most part, close to their average crustal abundance, and are not toxic at this level. Certain elements may be concentrated in coals at the top, bottom, or edges of a bed or basin (Ge, Be, Ga, B). Others may be enriched in samples close to mineral ore deposits (Mo, Sn, Zn, Pb, Hg, U). The potential danger from toxic elements such as Hg in coal has often been overstated by "averaging" results from mineral specimens collected in the search for ores. These conclusions are based on a compendium of the literature values for each element, by using the ranges and averages for each coal-producing region, after excluding atypical samples.*

*The volatile elements which are commonly considered most hazardous include F, As, Se, Cd, Pb, and Hg, on which the analytical data available have been spotty and unreliable. Improved methods for these analyses, which are now being developed, must be applied to both old and new samples.*

Geographical relationships of fuel source to quality have been useful in the selection of coals or crude oils for sulfur, ash content, and for many other properties. Any fuel specification limits supply and raises cost, and the addition of new specifications on trace elements could have important economic effects on cost and on availability. This paper examines some of the major variables in the amounts of trace elements in fossil fuels, and the extent to which geographical correlations may apply.

The literature is full of data on trace elements in coal or oil, but they are hard to use. Fuels are subject to all of the usual problems in sampling, and they have special ones of their own. Analyses for zinc in oil are useless if the sample is taken in a soldered can, and any unusual analysis for iron in coal is suspect. These problems of sample handling are typical, not rare. The problem continues from the field into the laboratory. There are classic examples of misleading results on mercury in coals where the whole laboratory was contaminated, and there remains the more recent discovery that laboratories in a downtown urban environment cannot get a good analysis for trace amounts of lead. The problem is not so serious in ordinary analyses down to about 0.1 to 0.01 percent, but the very words "trace element" refer to a situation where trace amounts of contamination are troublesome. A large number of elements in coal are present in trace amounts of about 1-1000 ppm. Cooperative tests in the round robin series conducted by the National Bureau of Standards have shown poor agreement between laboratories in the analyses of coals in the range of 1-50 ppm, and any analysis within this range requires rigorous calibration procedures, if the results are to be believed.

A more troublesome problem in the evaluation of literature data, because it is frequently ignored, is the basis for selection of the original sample. It is obvious that contamination is a problem for sodium chloride in crude oil, depending on how much salt water is left in the sample, or for adventitious clay minerals left in a coal. What is not so obvious is the effect that proximity to a zinc or tin deposit has on the amount of zinc or tin there is in coal.

Almost half of the coal produced is consumed as mined, without preliminary processing. The same is not true for petroleum, which is almost never burned as total crude. In both cases, any initial processing by cleaning or by fractionation usually produces a waste or bottoms fraction which is enriched in undesirable components, and this waste commonly creates pollution problems under present methods of handling or disposal. There problems are known and are fairly well defined for sulfur, but there is no comparable body of data for the pollution potentials due to other elements. A larger body of data is available for trace elements in coal. For both coal and petroleum, however, the level of trace elements present is so low that methods of sample selection and sample handling, prior to analysis, can and do present major complications in the interpretation of results.

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*This is a report of information obtained in a critical survey and analysis of the literature on trace elements as potential pollutants in fossil fuel conversion/treatment processes, under EPA Contract No. 68-02-0629. For details and bibliography, see contract report EPA R2-73-249, PB-225,039, June, 1973.

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Coal is a mineral resource, and most of the literature on coal analysis reflects its use as a guide to other minerals. In the first studies of uranium in coal, for example, the typical coal containing less than 10 ppm of the element was dismissed as of no interest, and data were reported for only the atypical exceptions. In later studies, a screening procedure was applied to select for trace metal analysis those samples which gave the highest percentage on ash of germanium, which was needed for electronics. This approach was explored during the 1950's and 60's in comprehensive reviews for a few elements such as uranium, beryllium, germanium, and gallium in the United States, and for mercury in the USSR. Concentrations of interest, from this point of view, cut off at about 10 ppm on ash or 1 ppm on coal, and few of the older analyses go below this level. An effort is made in this study to reduce this bias toward unusual specimens, and to draw correlations based on corrected data.

### *Evaluation of Data*

Continuing field surveys of U.S. coals have been made by three major government laboratories, using the best methods available to them. These surveys were made for three different purposes, and they differ more in the method of sampling than in the procedures for analysis. The rest of the literature on trace elements in coal is concerned almost entirely with new methods of analysis, tested or intercompared on a single sample or a few known samples. Except for missing data on single elements, these have relatively little to contribute to an overall review. The three basic sets of data and samples are:

(1) Surveys of commercial coals – U.S. Bureau of Mines (USBM) (delivered samples);
(2) Mineral source surveys – U.S. Geological Survey (USGS) (column samples);
(3) Specific element studies – Atomic Energy Commission (AEC) (TVA/Oak Ridge; USGS, specimens).

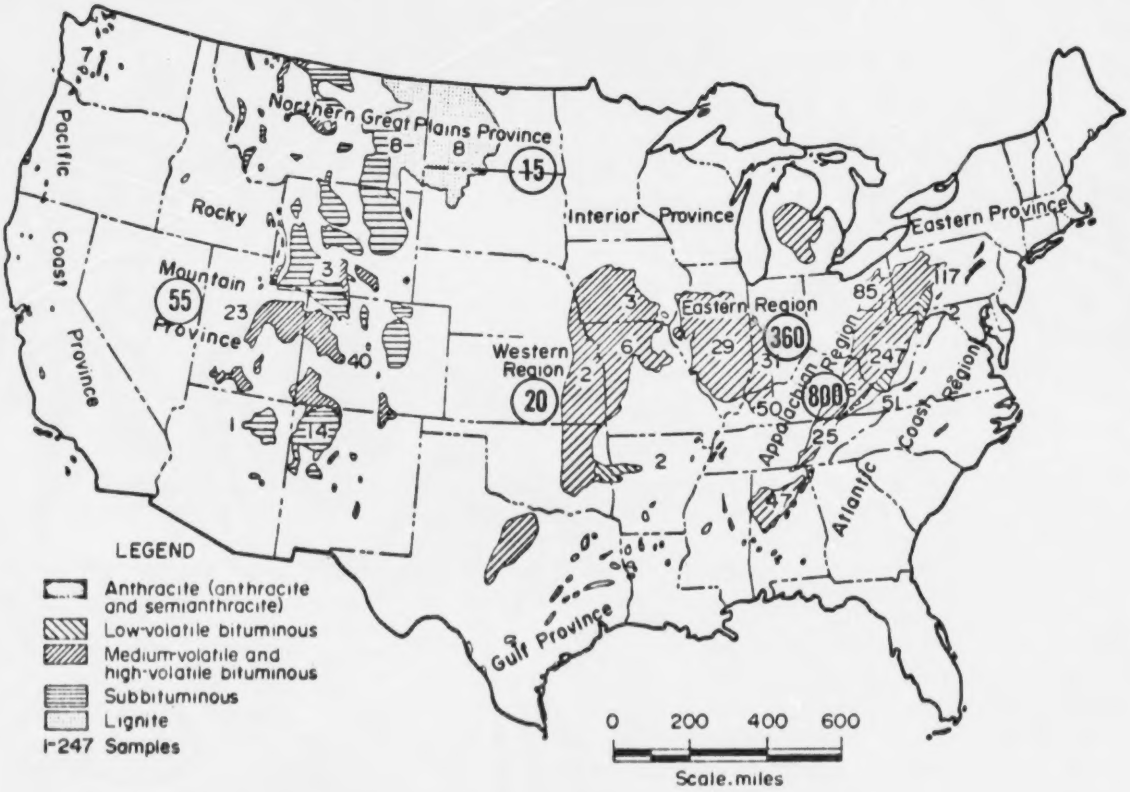
The USGS data, reported by Zubovic and Stadnichenko, are found to come closest to the purposes of the present program: to determine the composition of typical U.S. coals by regions, and the extent to which the selection of coals by geographic location can be expected to affect their composition in trace elements. The USBM has selected samples to cover current commercial production, and most of its data are reported on the basis of ash. The number of samples selected in the most comprehensive survey by the USBM in 1969 is shown by States and regions in figure 1, with superimposed numbers showing total production figures the same year for each region, in thousands of tons per day. This choice of samples is based on production rather than reserves and no distinction is made between cleaned and raw coals, which can make a significant difference in trace metals content.

The analyses made in these field surveys did not regularly include data on mercury, cadmium, arsenic, selenium, or fluorine. These are all volatile elements and are considered hazardous. They are not readily detected by emission spectroscopy, and better methods for their analysis were required.

Trace elements are usually defined as those elements that are present in the earth's crust to the extent of 0.1 percent (1,000 ppm) or less. Nearly all trace elements show an enrichment in coal ash relative to their crustal abundance. The USBM data on coal ash are summarized on this basis in figure 2. Boron picked up by plants from the underlying soil is enriched on the total coal basis (syngenetic), and germanium is equally enriched in some coals by exchange reactions during coalification (epigenetic). Manganese is low even on the ash basis. However, for most trace elements in coal, crustal abundance comes between the amount in coal and the amount in ash.

### *Potential Pollutants*

Public concern over minor or trace elements in coal as a potential source of air pollution was limited almost entirely to sulfur until recent years, after 1966. Data from USBM on sulfur in U.S. coals by States are shown in figure 3. These data show strong differences by geographical area, and they were discussed in detail in Congress when drafting the Clean Air Act of 1967. The initial concern and the first official actions were directed toward smoke control, to limit the production of soot, fly ash, and sulfurous emissions. The production of fly ash or cinders is linked with total ash content and composition, but it is scarcely affected by which trace elements are present in the ash.



Northern Great Plains Province
Pacific
Coast
Province
Rocky
Mountain
Province
Interior Province
Eastern Province
Eastern Region
Western Region
Appalachian Region
Atlantic Coast Region
Gulf Province
15
55
20
360
800
71
8
8
3
23
40
14
1
3
6
2
29
31
50
85
117
247
2
5L
25
47
2
LEGEND
Anthracite (anthracite and semianthracite)
Low-volatile bituminous
Medium-volatile and high-volatile bituminous
Subbituminous
Lignite
1-247 Samples
0
200
400
600
Scale, miles

Figure 1. Map Showing Number of Samples From Each State.

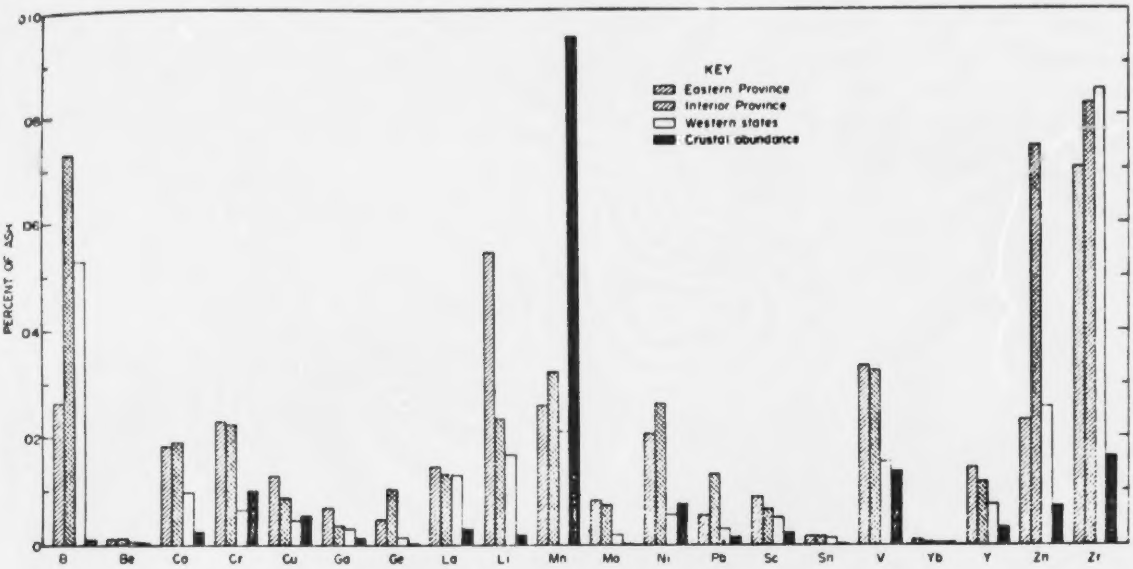


Figure 2. Average Trace Element Content in Ash of Coal From Three Areas Compared With Crustal Abundance.

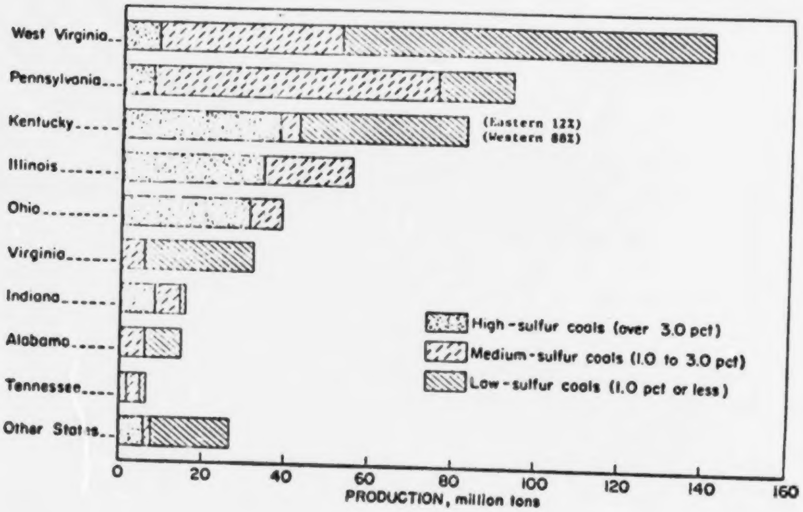


Figure 3. Total Coal Production of All Ranks, by State and Sulfur Content, in the United States, in 1964.

Coal rank, in general, is not a decisive factor in the relative amounts of different trace elements present. The coals of lowest rank, lignite and peat, show high ash with a very high content of calcium, sometimes of sodium, and barium in lignites, but the lignite ash contains so much alkali and alkaline earths that it is low in its content of other trace elements by simple dilution.

There are eight ash-forming elements, the components of common rocks or soils, which are usually present in coal ash to the extent of 1 percent or more. These elements, in the order of atomic weight, are: sodium, magnesium, aluminum, silicon, potassium, calcium, titanium, and iron. Their relative amounts may vary widely according to known principles of geology and the geochemistry of sedimentary deposits. Nitrogen is also present in all coals at 1 to 2 percent by weight, but the difference between one coal and another is too small to have much effect. These elements are not the subject of this report.

A list of the trace elements on which sufficient data are available for review is given in table 1. Three categories are shown, each in the order of increasing atomic weight:

The first category includes seven trace elements commonly considered as hazardous to human health.* It is undoubtedly no coincidence that these toxic elements are all volatile or can appear in stack emissions as volatile stable compounds. These elements as a group are hard to analyze, there are few data in the literature, and the data there are often

Table 1. Trace elements in coal (about 0.1 percent or less, on ash)

| Volatile, commonly considered hazardous | Others analyzed in coal (USGS) | Analyzed in ash (USBM) |
|---|---|---|
| Beryllium | Boron | Lithium |
| Fluorine | Vanadium | Scandium |
| Arsenic | Chromium | Manganese |
| Selenium | Cobalt | Strontium |
| Cadmium | Nickel | Zirconium |
| Mercury | Copper | Barium |
| Lead | Zinc | Ytterbium |
| | Gallium | Bismuth |
| | Germanium | |
| | Tin | |
| | Yttrium | |
| | Lanthanum | |
| | Uranium | |

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*This is in no sense an official list, but it does include all three of the elements (Be, Cd, Hg) named as hazardous to date by EPA.

unreliable. The second category includes trace elements analyzed in extensive field reviews and reported on a coal basis by Peter Zubovic and coworkers at the USGS. These were selected for its studies of the chemistry and geology of coals, and determined by a standard procedure for emission spectroscopy. Titanium, present regularly in amounts greater than 0.1 percent was not considered as a trace element in this review. The third category includes elements for which data are available only on an ash basis, from USBM studies of commercial coals.

### *Methods of Correlation*

Two factors have hampered efforts to draw useful correlations between the location of coals and their content of specific trace elements: the tendency to use averaged data and the fact that the basis of sampling for analysis has shifted during the course of major field surveys. The problem is particularly serious when, in the same program, samples are intermixed which are "of interest" for opposite reasons: because they are typical on the one hand, or because they are exceptional on the other. The analyst in this case finds no basis to discard a high result which he finds analytically correct, even though it greatly affects his average. This problem was recognized by the Geological Survey, for example, in the analyses reported for zinc in Illinois and elsewhere. This can be significant for pollution controls, since the expected analysis for delivered coals might be reduced to the mode by excluding a few samples which are easy to identify. The fact that such extremes do or do not exist and the producing areas in which they are found may be of definite interest in considering these trace elements as potential pollutants.

The unusual data should somehow be put into perspective so they can be used and not discarded, since the analyses involved are not in question. The literature was reviewed with these problems in mind. Three concepts are proposed as working tools to make the most out of the present literature, with a minimum of recalculations:

(1) A "variance" ratio is given for each element, which is the ratio of the highest to lowest average of analyses for areas or basins within the region. It is applied here to groups of about three States, as a measure of the extent to which selection of coal source can be expected to make a difference in the concentration of a given element. As shown in table 2, this variance is low for most elements, on the order of 2 to 3. It is 8 for sulfur and 4 or above for only 7 of the 18 elements for which sufficient data are available to report.

(2) Ranges for the USGS analysis of the coal beds within each region are selected which include 90 percent or more of the values reported, for columnar samples, up to a cutoff point above which higher values appear to be exceptional. The problem is that a sample cut from the mine face near the edge of a bed has a variable amount of bone, shale, or other minerals depending on how the sample is chosen. A columnar sample is defined here as one for which at least 75 percent of the total depth of the coal bed was included in the sample analyzed. This makes a significant difference in section A (1961) and B (1964) of the four-part USGS survey. At that time, the highest values reported were frequently not samples of straight coal but blocks where a major part of the sample was rejected as minerals before analysis. The 90+percent range is taken from the data for beds within the region, after casting out these nonrepresentative values.

(3) The next step involves inspection to find an envelope within which all or at least 90 percent of the individual analyses represent a continuous distribution of values. The highest values reported were then considered individually, first to see that they represent a columnar sample at least 75 percent analyzed, and then to determine the interval between this value and all lower values within the envelope. This inspection characterized many instances where a few columnar samples, up to about 5 percent, had extreme values at least 25 to 100 percent or more above the rest of the 90+percent envelope. The significance of the extremes can then be examined separately, both in terms of their frequency and their magnitude as compared to average values or the top of the 90+percent range. Where there are no extremes, the 90+percent envelope includes all samples. Detailed data on this are summarized in figures 4.1 to 4.7.

When the elements are listed in the order of variance and magnitude of extremes, in table 2, several useful correlations appear. The elements with high variance whose extremes are no more than twice the top of their 90+percent range are sulfur, germanium, boron, beryllium, and gallium. Except for sulfur, which is both strongly organic and inorganic, these are all at the top of the USGS list of organic affinity. This means that on coal cleaning they tend to be associated with the coal fraction, and not with the waste. They are also elements in which coal is greatly enriched, to the extent that it is

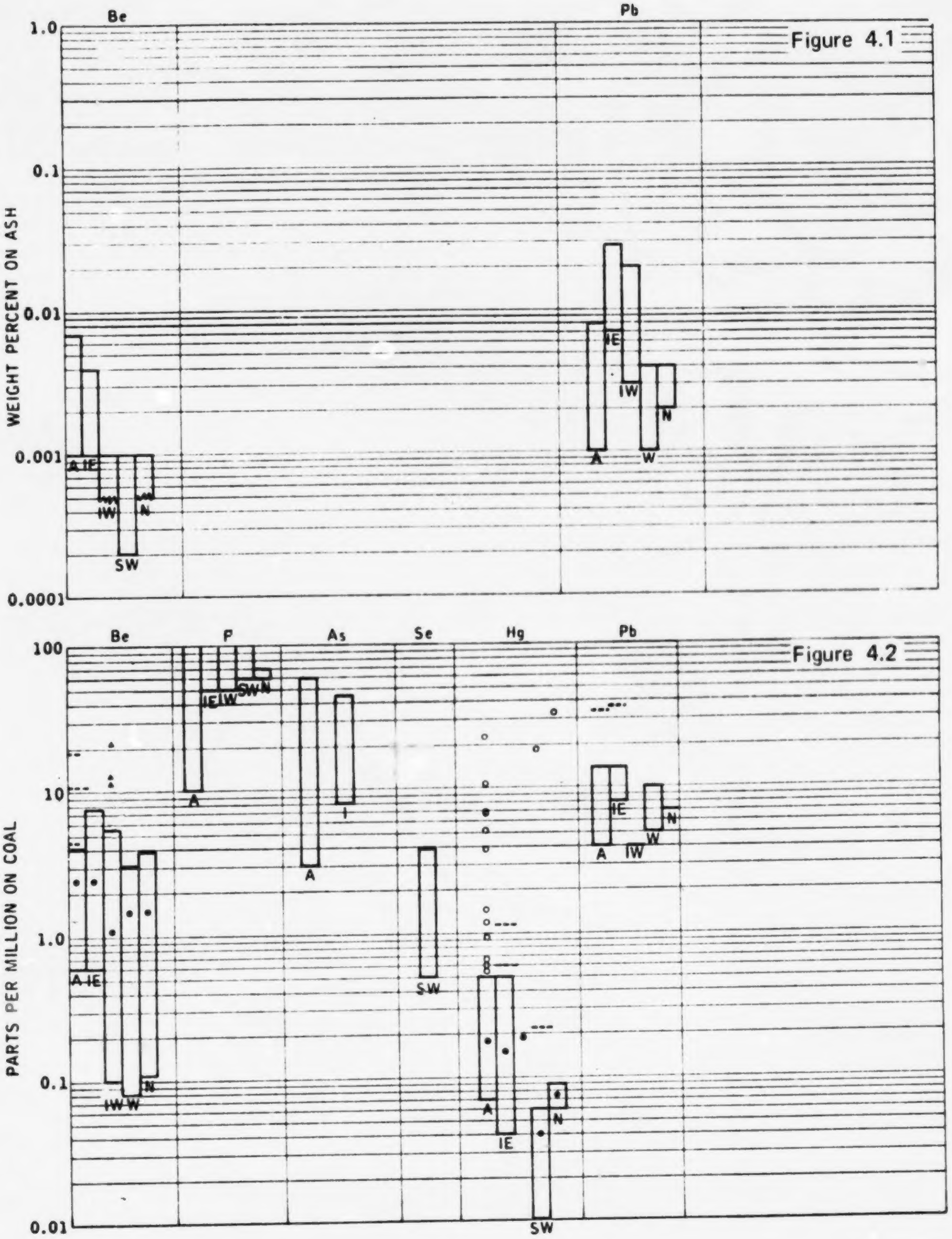
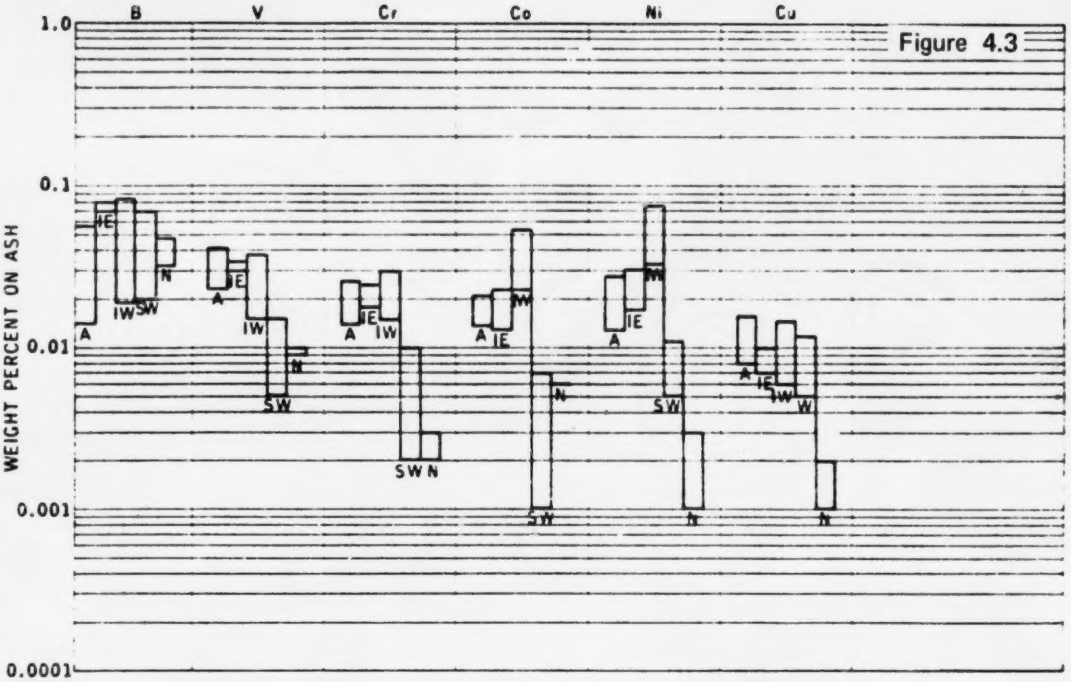


Figure 4a. TRACE ELEMENTS IN U.S. COALS



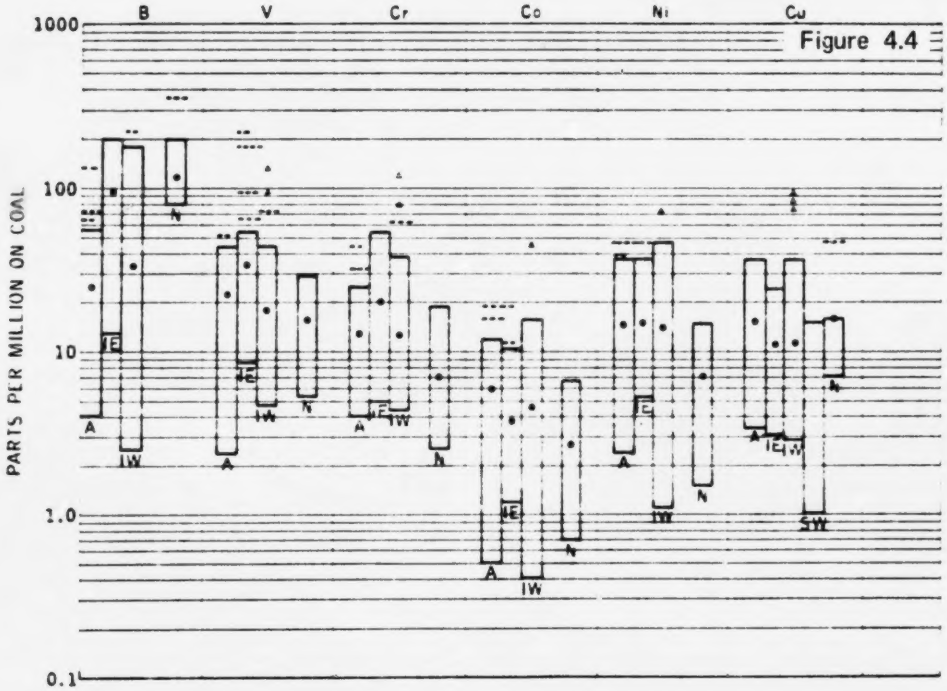
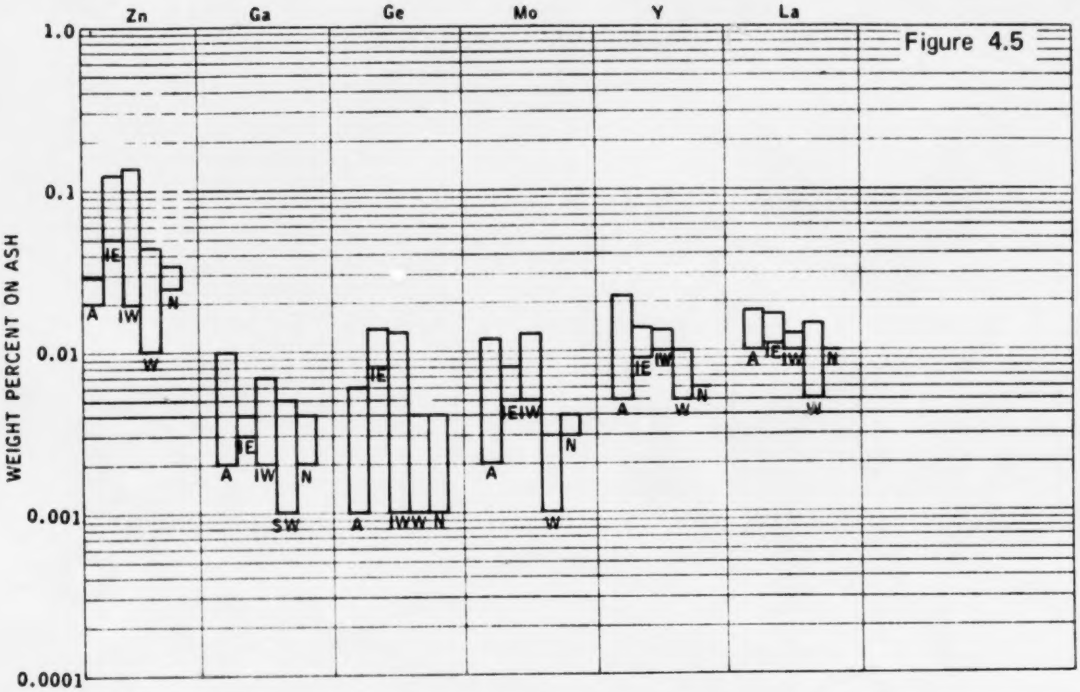


Figure 4b. TRACE ELEMENTS IN U.S. COALS



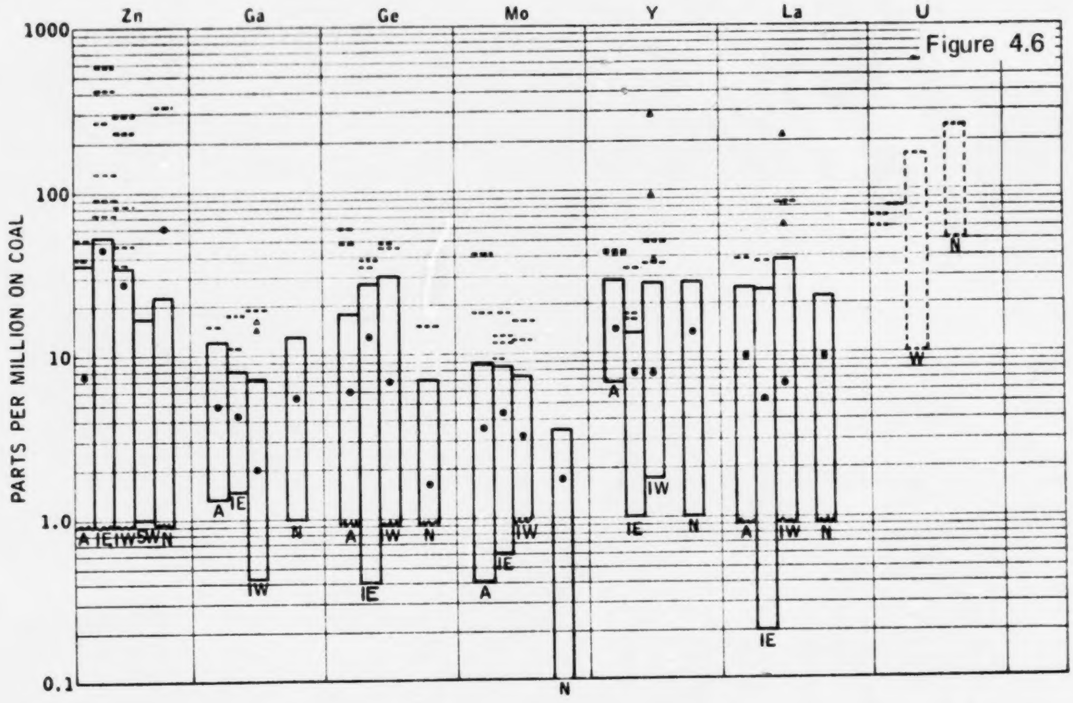


Figure 4c. TRACE ELEMENTS IN U.S. COALS

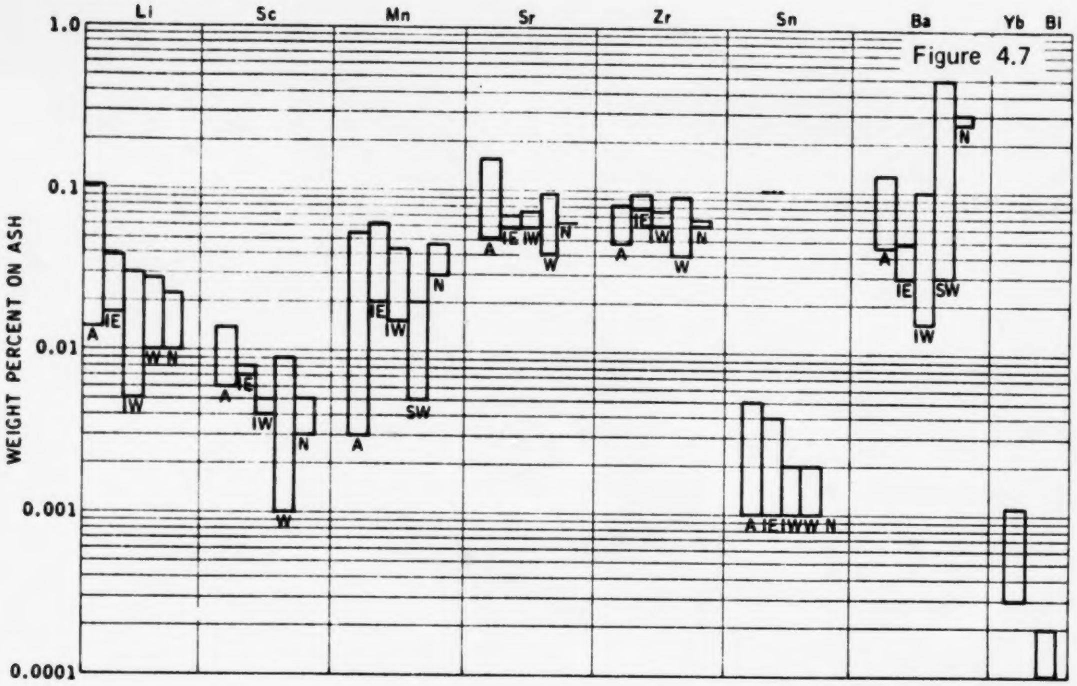


Figure 4.7

Figure 4d. TRACE ELEMENTS IN U.S. COALS

recognized for most of them as a significant mineral source.

The other elements which show a variance above 4 are zinc, molybdenum, and tin. For these, the extremes reported are from 5 to 20 times the top of the 90+percent range. Zinc and tin are at the bottom of the organic affinity list, and for them the occurrence of coals of high metal content is taken as a direct indication that ore beds are nearby. The variance of (3) reported for tin is placed in parenthesis because it represents a recalculation of the USGS data for one area in the Appalachian region. This is given a false low average of 0.1 in the original report by including a value of zero for 15 of the 17 samples analyzed for the region, in which the element was not detected. The USBM data in this area indicate no such differences between the tin content of coals in this area and in other parts of the region, so this value was ignored in calculating the variance.

Variance relates to geography, and extremes relate to mineral enrichment. Molybdenum is another case where variance alone might be misleading. Here the average for the southern Appalachian area, Alabama, is raised from 3.9 to 5.8 by a single extreme value of 42 for one bed out of the 20 included. Without this one extreme, the variance reported would be under 3. The value appears valid but marginal (74 percent of bed analyzed), and the fact that it is so high may correlate directly with the AEC observation that Mo is the only element whose occurrence in coal in high amounts they could link with the occurrence of high amounts of uranium.

Mercury and lead, for which no data are reported in the USGS field survey, have a variance of about 2 to 3, using other data. The ratio of extremes to average values for these two elements can be given any high value desired (up to 50-100) depending on how close one chooses to come to an ore bed in selecting the coal. None of the remaining elements which show a variance of 3 or less has a ratio of extremes to the top of the 90+percent range higher than 2, except vanadium at 3. This vanadium ratio directs attention to one mine in Kentucky, which has a high Ni and V content.

The correlation concepts of variance and extremes were developed element by element for the 5 major producing regions: Appalachian (A), Interior-Eastern (IE), Interior-Western (IW), Western (W), and Northern Plains (N). The data summarized in table 3 are presented in figures 4.1 to 4.7 as bar charts for each element, by regions, following the listing of elements in table 1. In some cases where the Southwest Energy Study appears to give a broader data base than the W ranges, based on more samples, SW data have been reported instead of W. The USBM data for ppm on ash are shown at the top, and the USGS data on a coal basis are at the bottom. Thousandths of a percent on ash correspond to ppm on coal, if the coal contains 10 percent of ash. This is a reasonable average approximation, and it fills in many holes in the data without a laborious recalculation, which would still not give directly comparable results.

The bar graphs shown for coal are the 90+percent ranges, the dashed lines (---) are the extremes listed, and the regional average (°) is for the total region as given by USGS. This average is usually near the middle of the bar, but it moves toward the top of the bar or may exceed it when there are many extremes, as for copper or zinc. Ranges which start below the limit of detection are shown by a broken bar line below 0.1 ppm. Shorter dashed lines (--) represent values outside the 90+percent range which were included in the USGS average, but are excluded here because they were for beds less than 75 percent analyzed. High specimen sample values for mercury reported in the literature are indicated by an o, and Δ shows the high values for weathered samples, not included in the averages.

The bar graphs for most elements, thus adjusted, lie within the range of 1 to 50 ppm, and most are close to 5-10 ppm on coal. The only elements significantly higher than this are boron and fluorine, in the range from 10 to 200 ppm. Beryllium is lower in all regions by an order of magnitude, at about 0.1 to 5 ppm, and Hg by two corders of magnitude, at about 0.01 to 0.5 ppm on coal.

*Mercury: Averages and Extremes*

The literature bias toward samples of mineral interest must be discounted heavily in certain cases. A specific example is mercury, which has received major attention in the past 2 years as a hazardous air pollutant. Concerted efforts have been made to supply missing data on its analysis in fossil fuels. Part of this concern is based on the widely quoted statement that the amount of mercury released to the environment by the burning of coal is comparable to that emitted as waste from all industrial processes. This statement is taken from a 1971 article in which 11 of 36 specimens analyzed contained over 1 ppm of Hg on coal, but 8 of the 11 were samples from the same three counties. The author of this article states

Table 2. Variances and extremes in areas[a]

| Element | Variance between areas[b] | Organic affinity[c] | Extreme ratio[d] |
|---|---|---|---|
| Sulfur | 8 | | |
| Germanium | >10 | 1 | 2 |
| Boron | 6 | 3 | 2 |
| Beryllium | 5 | 2 | 2 |
| Gallium | 4 | 3 | 2 |
| Molybdenum | 4 | 6 | 5 |
| Tin | >(3) | 8 | 5 |
| Zinc | >5 | 10 | 20 |
| Lead | 3 | | >10 |
| Mercury | 3 | | >10 |
| Vanadium | 2 | 3 | 3 |
| Nickel | 3 | 4 | 2 |
| Chromium | 2 | 4 | 2 (3)[e] |
| Cobalt | 2 | 5 | 2 (5) |
| Yttrium | 3 | 5 | 2 (10) |
| Copper | 2 | 7 | 2 (3) |
| Lanthanum | 3 | 9 | 2 (10) |

[a] From USGS bulletin 1117, except for S, Pb, and Hg.

[b] Approximate ratio between averages for highest and lowest areas, as published (see PB-225 039, table XI).

[c] Numbers assigned to USGS order of affinity: Ge>Be>(Ga,Ti,B,V)>(Ni,Cr)>(Co,Y)>Mo>Cu>Sn>La>Zn

[d] Approximate ratio between extreme and top of the range of 90+ percent envelope, for columnar beds analyzed.

[e] Discounted extremes in parentheses show exaggerated effect of two weathered coals from Arkansas with 41.7 and 47.3 percent ash, not included in area averages.

that, "The analyses were performed on a relatively small number of samples that are not representative," but this qualification has been completely lost in subsequent references. He finds that his method of analysis is confirmed by agreement between his average of 0.19 ppm for coals from Illinois (5 samples) and the average value of 0.18 ppm reported by the Illinois State Geological Survey (53 samples). He then recognized that his sampling of 36 U.S. coals gives an average which is too high (3.3 ppm), but apparently gives his extreme values equal weight with the samples determined elsewhere and chooses a "conservative estimate" of 1 ppm as typical of all coal produced. On this basis, he calculates 3,000 tons of mercury per year are released by coal combustion, worldwide, and finds this quantity comparable to the 10,000 tons per year consumed industrially, most of which is eventually discarded to waste. Thus, this is the origin of the statement so widely quoted. The more representative basis of 0.18 ppm as typical for coals would give 540 tons of mercury from combustion stacks as against 10,000, which is less than "comparable" by an order of magnitude.

Comprehensive studies of U.S. coals during 1971 and 1972 have failed to find a single commercial supply which runs as high as 1 ppm. The newer methods of analysis have a limit of detection of about 0.01 ppm. This is approximately the same by neutron activation and by flameless atomic absorption, using a double gold amalgamation procedure to remove interferences without loss of mercury. Data which permit a good survey of geographical distribution by producing region are presented in table 3, and a proper U.S. average might be close to 0.15 ppm.

The reason for this confusion lies not in the first author's method of analysis, which is reliable, but in the selection of samples which are essentially museum specimens, and in allowing them to be included in a result reported as "average." The present study fully confirms on a smaller scale the general observation that the mercury content in most coals is quite low, less than 0.2 ppm, and that occasional extremes as high as 1 ppm are limited to a few specific locations. Even where these extremes occur, the average Hg content for the mine is usually far below the extreme, and is not necessarily much different from the average for other coals.

### *Problems in Toxicity*

The problem of identifying what is toxic is, in many cases, an open question. Biological evolution is a rigorous adaptation of the organism to its environment, and a large number of trace elements are essential to life as we know it. The base value for toxicity is not zero, but close to "crustal abundance," and too little of these elements is as toxic as too much. It should be recognized, however, that this is as true for common foods as it is for trace elements. The body cannot tolerate sodium chloride or water at 10 times its normal intake. The question of toxicity is tied closely to actual amounts, and the tolerance range between food and poison is not wide. Diet deficiencies due to overuse of the soil are well known for many elements, and the addition of coal ash to the environment may be beneficial to plants or to people whose food and living space have been deprived of contact with virgin soil.

A useful overview of trace elements in rocks, soils, plants, and humans and the known facts of their toxicity has been assembled by Lisk. His averages are shown in table 4 together with ranges for ppm in coal taken from the present review. This comparison makes even more striking the number of trace elements in coal which show about the same concentration, in the range of 1 to 50 ppm. The amounts of different trace elements in coal are apparently more uniform than they are in rocks or soil. The human body is relatively more rich in lead, mercury, and cadmium than in coal, and the same applies to barium, strontium, and lanthanum. Coalification is a geologic averaging process, and the human body has different selection principles, which depend at least partly on atomic weight. Enrichment in plants follows still a different set of principles. For reasons which are not clear, plants seem to selectively reject (enrichment below 0.03) two groups of elements having adjacent atomic numbers: scandium, titanium, vanadium, chromium (21 to 24), antimony, and tellurium (51, 52). Elements which plants do select include cadmium, lead, silicon, selenium, and lithium.

Elements which are considered toxic have a tendency toward biologic methylation. This is true for arsenic, mercury, and selenium. This is partly a matter of valence state. The difference between calomel and corrosive sublimate is well known, and toxicity depends heavily on chemical form for beryllium or fluorine. Many toxic elements form stable chelates: vanadium$^{5+}$, zirconium$^{4+}$, mercury, thallium, and lead.

Toxicity involves many metabolic interactions. Arsenic, for example, may be an essential element at soil levels, up to 40 ppm. It counteracts certain diet deficiencies and the effects of excess selenium.

Table 3. Geographical distribution of mercury
1971-72 results; PPM on coal

| | | Analysis by | | | | |
|---|---|---|---|---|---|---|
| Region | State | Neutron activation (Illinois)[a] | N.A. + AA (N.B.S.)[b] | Flameless -AA (USGS-SW)[c] | Average | Total No. of Samples |
| A | Pennsylvania | 0.16,0.28 | 0.15 | - | 0.20(2.0[d]) | 3 |
| | Ohio | 0.10,0.13,<br>0.15 | 0.14,0.28,<br>0.49 | - | 0.21 | 6 |
| | West Virginia | - | 0.07,0.18 | - | 0.12(6.6[d]) | 2 |
| | E. Kentucky | - | - | - | (0.25[d]) | - |
| IE | Illinois | 0.04,0.49,<br>0.60,1.15 | - | - | 0.18(0.19[d]) | 53 |
| | Indiana | - | 0.08 | - | 0.08(0.31[d]) | - |
| IW | Missouri | - | 0.19 | - | 0.19 | - |
| N | Montana | 0.06 | 0.07,0.09 | - | 0.07,(33.0[d]) | - |
| W | Utah | 0.04 | - | 0.03-0.08 | 0.05 | 15 |
| | Colorado | 0.02,0.02 | 0.05 | 0.03-0.06 | 0.04(0.22[d]) | 3 |
| | Wyoming | - | - | 0.03-0.06 | 0.05(18.6[d]) | 6 |
| | Arizona | 0.02 | 0.06 | 0.04-0.08 | 0.05 | 6 |
| | Nevada | - | - | 0.04-0.05 | 0.05 | 7 |
| | New Mexico | - | - | 0.05-0.29 | 0.15 | 37 |

[a] Illinois State Geological Survey, bulletin EGN-43, 1971.

[b] National Bureau of Standards, 1972.

[c] Southwest Energy Study, appendix J, (draft), Jan. 1972.

[d] Values from Joensuu (1971) shown for comparison, including lithotypes; extremes show no relationship to more representative average samples.

Table 4. Trace elements in soils, plants, and animals[a]

| | Rocks[b] (ppm) | Soil (ppm) | Ratio plants/soil | Human body (mg total) | Coals (ppm range) 1.50 | 0.2-10 |
|---|---|---|---|---|---|---|
| As | 1-3 | 6 | 0.03 | 7.9 | x | |
| Ba | 50-580 | 500 | 0.11 | 22 | x(+) | |
| Be | 1-6 | 6 | 0.03 | 0.04 | | x |
| Cd | 0.03-0.3 | 0.06 | 5.3 | 50 | | ? |
| Cr | 35-100 | 100 | 0.01 | 1.7 | x | |
| Ga | 12-19 | 30 | 0.04 | | | x |
| Ge | 0.8-5.4 | 1 | 0.25 | | x | |
| Pb | 7-20 | 10 | 2.3 | 120 | x | |
| Hg | 0.03-0.4 | 0.3 | 0.05 | 13 | | x(-) |
| Ni | 2-75 | 40 | 0.05 | 10 | x | |
| Se | 0.05-0.6 | 0.2 | 1.0 | 13 | x | |
| Sr | 20-375 | 300 | 0.09 | 320 | x(+) | |
| V | 20-135 | 100 | 0.008 | 48 | x | |

[a] Donald J. Lisk, Advances in Agronomy, Vol. 24, pp. 267-324 (Academic Press, 1972). Data on coals from present report.

[b] Average values for igneous rocks, shales, and sandstones.

Selenium is essential to plant and animal life. It is specific to some plants, rarely deficient in man, and protects against excess cadmium or mercury. On the other hand, with selenium there is only a narrow range between deficiency and toxicity. Chromium is essential for glucose metabolism, copper is essential to enzymes, and cobalt and molybdenum are essential in blood. Even barium and strontium may be essential, as shown by a British survey which found uniformly high amounts in the tissues of partridge taken in areas where the hunting was good.

Further correlations of this type are still to be made, and the question of what is toxic to humans is not the same as for other organisms.

### *Conclusions*

1. Overall, geography is an important factor in distribution for only a few trace elements in coals. Many elements are present at about 1 to 100 ppm in all regions, and vary by a factor of 3 or less in the averages for different basins or areas. The usual amount of some 20 trace elements measured is about 5 to 10 ppm. B and F are higher, at about 50 to 200 ppm; Hg is lower, about 0.01 to 0.5 ppm.
2. The volatile hazardous elements show the most need for more data. Some or all of these may be related to mineral deposits nearby (Be, F, As, Se, Cd, Hg, Pb). The hazard of Hg in power plant emissions, in particular, has been grossly exaggerated.
3. Most elements in coal are very close to their crustal abundance, and are not considered toxic at this level. The question of toxicity must be evaluated in relation to actual amounts.
4. The few elements which tend to be concentrated in coals (S, Ge, Be, B, Ga) are associated primarily with the organic portion of the coal. They also show the largest variance in average concentrations between different producing areas, e.g., for germanium, which is high in Illinois.
5. The amount of some trace elements is commonly highest in the top and in the bottom few inches of a bed, and at the edges of a coal basin (Ge, Be, Ga, and B at bottom only). These variations are frequently greater than the differences between the averages for different beds. Other elements (Cu, Ni, Co) shown no such correlation.
6. Anomalous amounts of specific elements may be found in beds contiguous to mineral ore bodies of the same element. This is regularly the case for coals having a mercury, lead, zinc, or uranium content higher than the usual range, and may be equally true for other elements including copper, tin, and arsenic.
7. The elements present in the largest amount, as minor components of the coal rather than as traces only, are the common constituents of surface waters: silica, alumina, iron, sulfur, phosphorus, sodium, potassium, calcium, and magnesium. These are present throughout the coal but they are often enriched in the top layer, where they have apparently been leached out of enclosing sediments.
8. The selection of a completely "nonpolluting" coal is not possible, in the general case. For a given amount of ash, coals which are low in any one group of elements must be correspondingly high in others. The definition of nonpolluting depends directly on the decision as to which elements are of concern and which are not.

# DISTRIBUTION OF TRACE ELEMENTS IN COAL

R. R. Ruch, H. J. Gluskoter, and N. F. Shimp*

## *Abstract*

*Complete analyses of 101 coals, mostly from Illinois and a few from eastern and western U.S. coal fields, have been made in the laboratories of the Illinois State Geological Survey. Specific gravity separations up to 2.89 were made on four representative coals, and subsequent analyses were performed on the 33 float-sink fractions resulting from the separations. Trace elements determined were Sb, As, Be, B, Br, Cd, Cr, Co, Cu, F, Ga, Ge, Pb, Mn, Mo, Ni, Hg, P, Se, Sn, V, Zn, and Zr. Major and minor elements Ca, Cl, Fe, Mg, K, Na, Si, S, and Ti.*

*The analytical techniques employed were neutron activation, optical emission, atomic absorption, X-ray fluorescence, and ion-selective electrode. Accuracy and precision were maximized by interlaboratory and intralaboratory comparisons. High-temperature ash (~ 500°C), low-temperature ash (< 150°C), and whole coal samples were analyzed in this investigation. Trace element volatilities were determined for each sample type, and further analyses were done on the samples which best retained the element to be determined.*

*Concentrations of some trace and minor elements varied widely according to areal and stratigraphic distribution of the samples. Significant statistical correlations of a number of elements (e.g., Zn and Cd; Si, Ti, Al, and K; Ca and Mn; and Co, Ni, As, Cu, and Pb) are readily attributable to the mineral phases present.*

*Association of some elements with inorganic mineral fractions in the coals is further demonstrated by their concentrations in the heavier specific gravity fractions of the float-sink samples. Washability studies indicate the potential effective removal of several trace elements (including Zn, Cd, and Pb) from raw coals by conventional specific gravity methods. Other elements (including B, Be, and Ge) are not removed by these methods because a large portion of each is in organic combination.*

*The presence in coals of Zn and Cd in the mineral sphalerite, of Pb in galena, of P and F in apatite, and Ni and Cu in the iron sulfides has been demonstrated by scanning electron microscopy and energy dispersive X-ray analyses.*

*This study demonstrated the various problems of a relatively complete coal analysis, including sampling, sample preparation, adaptation of specific analytical procedures, and interpretation of the statistical treatment of the data.*

## INTRODUCTION

It has only recently become apparent that a thorough knowledge of trace elements in coal is necessary. Some trace elements in coal such as arsenic, beryllium, cadmium, mercury, and lead are known to be toxic to plant and animal life at low concentrations. The problem potentially extends beyond volatilization to the long-term storage of fly ash and bottom ash. In addition, some elements may act as catalysts or may retard catalysis in proposed coal-conversion processes. Hence, accurate and reliable data must be developed concerning the absolute amounts of these elements in coal, their distribution and modes of occurrence, and their volatility during combustion.

Several recent review articles deal with the subject of the chemical nature of coal, including trace and minor elements (refs. 1-4); however, most analyses prior to 1970 were done on high-termperature ash and do not reliably indicate the total amount of volatile trace elements in whole coal, e.g., Hg, Br, F, etc. This study concentrates not only on determining trace elements in whole coal but also on determining their volatility when the coals are plasma-ashed at low temperatures (< 150°C) and when conventionally ashed at high temperatures (up to 700°C). A series of prepared coals was also analyzed to determine which elements might be reduced by specific gravity cleaning techniques.

This short report is a much abridged condensation of a manuscript currently being prepared for publication by the Illinois State Geological Survey as an Environmental Geology Note (EGN). The research reported upon here and in the forthcoming EGN was supported, in part, under Contract No. 68-02-0246 and Grant No. R-800059 from the U.S. Environmental Protection Agency, Demonstration Projects Branch, Control Systems Laboratory, Research Triangle Park, North Carolina. An interim report on this project has also been published (ref. 5).

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*Experimental*

Complete detailed information about sampling, sample preparation, chemical analysis, and data treatment is presented in other reports (ref. 5, and in the final report, in preparation). Only a brief summary is possible here.

Three face-channel samples were collected from each mine tested, combined into a composite sample, crushed to pass a 1/8-inch screen, riffled, and comminuted to minus 20 mesh, minus 100 mesh, or finer, depending upon the analytical technique to be used. It was found, through X-ray fluorescence studies, that minus 200 mesh is optimum for acceptable precision.

Samples of whole coal, low-temperature ash (LTA, < 150°C, derived from radio-frequency, oxygen-plasma ashing of whole coal), and high-temperature ash (HTA, ~ 500°C in a muffle furnace) were all used for chemical analysis, and the results were compared. Volatility studies were conducted to ascertain which elements might be lost, wholly or in part, under the different ashing conditions employed. A summary of these results is shown in table 1.

The volatility results dictate that Hg, Br, Sb, and F must be determined on whole coal; whereas Ga, Se, As, Zn, Ni, Co, Be, Cu, Pb, V, Mn, and Cr can all be determined on the low-temperature ash. High-temperature ash is suitable for analyses of Zn, Ni, Cr, Cu, Pb, B, Cd, Mn, Cr, Be, Ge, and Sn. Indications are that Mo and possibly V are partially volatilized at 500°C.

Analytical techniques employed were neutron activation analysis (NAA), atomic absorption (AA), optical emission-direct reading (OE-DR), optical emission-photographic (OE-P), X-ray fluorescence (X-RF), and ion-selective electrode (ISE).

Where possible, extensive cross-checking was conducted. This included interlaboratory and intralaboratory comparisons, including participation in a roundrobin program on coal analysis cosponsored by the EPA and the NBS. Accuracy and precision were emphasized over speed and were used as the criteria for choice of technique. Table 2 summarizes the various techniques chosen and the types of samples used for analyzing the various elements.

In general, individual precision for each element is 15 percent or less, with sensitivities of about 1 to 10 ppm on the whole coal basis.

## DISCUSSION

Complete chemical analyses of 101 whole coal samples and of 32 laboratory-prepared samples obtained by specific gravity separation of four coals have been made in the laboratories of the Illinois State Geological Survey. Of the 101 whole coal samples, 82 were from the Illinois Basin (Illinois, Indiana, and western Kentucky). The additional 19 samples were from other areas of the United States.

Table 1. Summary of trace element volatility losses in pretreatment ashing of coal

| Low-temperature ash | | | High-temperature ash | | |
|---|---|---|---|---|---|
| Retained (> 95%) | | Lost | Retained* | | Lost |
| Ga | Cu | Hg (up to 90%) | Zn | Cd | Mo (33%) |
| Se | Pb | Br (100%) | Ni | Mn | V (up to 25%) |
| As | V | Sb (up to 50%) | Co | Cr | |
| Zn | Mn | F (untested) | Cu | Be | |
| Ni | Cr | | Pb | Ge | |
| Co | Cd | | B | Sn | |
| Be | | | | | |

*No significant losses observed in coal ash from 300° to 700°C or no significant differences observed between results from whole coal to low-temperature ash and high-temperature ash (~ 450°C).

Table 2. Analytical procedure recommended

| Element | Sample | Procedure |
|---|---|---|
| Hg.Mn,Na | Whole coal | NAA |
| Sb,Se,As, Ga | LTA | |
| Fe,Ti,Al, Si,K,Ca, S,Cl,Mg, Br,P | Whole coal | X-RF |
| Be,Ge,Co, Cr | HTA | OE-DR,OE-P |
| Cd,Zn | LTA | AA |
| Pb,Cu | LTA<br>HTA | AA<br>OE-DR |
| Ni | HTA<br>LTA<br>Whole coal | OE-DR,OE-P<br>AA<br>X-RF |
| F | Whole coal | ISE |
| Zr | HTA | OE-P |
| V | Whole coal,LTA<br>HTA* | X-RF<br>OE-DR,OE-P |
| Mo,* Sn,B | HTA | OE-DR |

*Low result is possible because of volatilization.

The 4 coals which were prepared in the laboratory (washed) were also from the Illinois Basin. Trace elements determined were antimony, arsenic, beryllium, boron, bromine, cadmium, chromium, cobalt, copper, fluorine, gallium, germanium, lead, manganese, mercury, molybdenum, nickel, phosphorus, selenium, tin, vanadium, zinc, and zirconium. In addition, the following major and minor elements were also determined: aluminum, calcium, chlorine, iron, magnesium, potassium, silicon, sodium, sulfur, and titanium. Standard coal analyses-i.e., proximate, ultimate, heating value, varieties of sulfur, and ash--also reported.

As the first steps in the statistical analyses of the more than 6,000 analytical values generated, the arithmetic means, standard deviations, ranges, and linear correlation coefficients were calculated for the trace and major elements, for the high- and low-temperature ashes, and for the proximate and ultimate coal analyses for the 101 coals tested (table 3).

On the basis of these statistical calculations and histograms of the element distributions, the elements can be grouped with those of similar type. In the first group of elements, each displays a relatively normal distribution of analytical values and has small standard deviations and ranges. Included in this group are Al, Fe, F, Ga, Be, Br, B, Cr, Cu, K, Ni, Si, Ti, Se, and V. In the second group of elements, each has a skewed pattern of analytical values with large standard deviations and ranges. This group includes Cd, Zn, P, As, Sb, Pb, Sn, Cl, Ge, and Hg. The first group of elements includes many with organic affinities. Some of these elements are thought to be syngenetic, and therefore inherited from an early period of coal swamp formation. The second group includes elements commonly found in coal, and in sedimentary rocks in general, as carbonates and sulfides. These minerals are often emplaced in coal by epigenetic mineralization.

Correlation coefficients for the various parameters determined for the coals from the Illinois Basin and for the entire 101 coals demonstrate the following geochemical associations:

1. The highest value for the correlation coefficients determined is that between Zn and Cd ($r = 0.93$). Both zinc and cadmium are present in coals in the mineral sphalerite, and probably only in that form.

2. The "Chalcophile" elements are those elements commonly found in nature as sulfides; they include As, Co, Cu, Ni, Pb, and Sb, which are all positively correlated with each other in the coals analyzed.

3. The lithophile elements, those commonly occuring in nature as silicates, include K, Ti, Al, and Si, which also have mutual positive correlations in the data reported. These elements are found in coals primarily as clay minerals (aluminosilicates).

4. There is a positive correlation of 0.63 between Mn and Ca in the coals analyzed, and Mn does not correlate as well with any other parameter. Mn is present in small amounts and most likely is in solid solution with Ca in calcite ($CaCO_3$).

5. Sodium and Cl have a positive correlation of 0.53 in the coals studied.

Several additional geochemical relationships have been suggested by the chemical analytical data, such as:

I. The concentrations of As, Cu, Pb, Si, and Al are lower in the younger coals of the Illinois Basin than in the older coals.

2. The correlation between Na and Cl increases

## TABLE 3. Mean Analytical Values for All 101 Coals

| Constituent | Mean | Standard Deviation | Minimum | Maximum |
|---|---|---|---|---|
| As | 14.02 ppm | 17.70 | 0.50 | 93.00 |
| B | 102.21 ppm | 54.65 | 5.00 | 224.00 |
| Be | 1.61 ppm | 0.82 | 0.20 | 4.00 |
| Br | 15.42 ppm | 5.92 | 4.00 | 52.00 |
| Cd | 2.52 ppm | 7.60 | 0.10 | 65.00 |
| Co | 9.57 ppm | 7.26 | 1.00 | 43.00 |
| Cr | 13.75 ppm | 7.26 | 4.00 | 54.00 |
| Cu | 15.16 ppm | 8.12 | 5.00 | 61.00 |
| F | 60.94 ppm | 20.99 | 25.00 | 143.00 |
| Ga | 3.12 ppm | 1.06 | 1.10 | 7.50 |
| Ge | 6.59 ppm | 6.71 | 1.00 | 43.00 |
| Hg | 0.20 ppm | 0.20 | 0.02 | 1.60 |
| Mn | 49.40 ppm | 40.15 | 6.00 | 181.00 |
| Mo | 7.54 ppm | 5.96 | 1.00 | 30.00 |
| Ni | 21.07 ppm | 12.35 | 3.00 | 80.00 |
| P | 71.10 ppm | 72.81 | 5.00 | 400.00 |
| Pb | 34.78 ppm | 43.69 | 4.00 | 218.00 |
| Sb | 1.26 ppm | 1.32 | 0.20 | 8.90 |
| Se | 2.08 ppm | 1.10 | 0.45 | 7.70 |
| Sn | 4.79 ppm | 6.15 | 1.00 | 51.00 |
| V | 32.71 ppm | 12.03 | 11.00 | 78.00 |
| Zn | 272.29 ppm | 694.23 | 6.00 | 5,350.00 |
| Zr | 72.46 ppm | 57.78 | 8.00 | 133.00 |
| Al | 1.29 % | 0.45 | 0.43 | 3.04 |
| Ca | 0.77 % | 0.55 | 0.05 | 2.67 |
| Cl | 0.14 % | 0.14 | 0.01 | 0.54 |
| Fe | 1.92 % | 0.79 | 0.34 | 4.32 |
| K | 0.16 % | 0.06 | 0.02 | 0.43 |
| Mg | 0.05 % | 0.04 | 0.01 | 0.25 |
| Na | 0.05 % | 0.04 | 0.00 | 0.20 |
| Si | 2.49 % | 0.80 | 0.58 | 6.09 |
| Ti | 0.07 % | 0.02 | 0.02 | 0.15 |
| Org. S | 1.41 % | 0.65 | 0.31 | 3.09 |
| Pyr. S | 1.76 % | 0.86 | 0.06 | 3.78 |
| Sul. S | 0.10 % | 0.19 | 0.01 | 1.06 |
| Tot. S | 3.27 % | 1.35 | 0.42 | 6.47 |
| SXRF | 2.91 % | 1.24 | 0.54 | 5.40 |
| ADL | 7.70 % | 3.47 | 1.40 | 16.70 |
| Mois. | 9.05 % | 5.05 | 0.01 | 20.70 |
| Vol. | 39.70 % | 4.27 | 18.90 | 52.70 |
| Fix. C | 48.82 % | 4.95 | 34.60 | 65.40 |
| Ash | 11.44 % | 2.89 | 2.20 | 25.80 |
| Btu | 12,748.91 | 464.50 | 11,562.00 | 14,362.00 |
| C | 70.28 % | 3.87 | 55.23 | 80.14 |
| H | 4.95 % | 0.31 | 4.03 | 5.79 |
| N | 1.30 % | 0.22 | 0.78 | 1.84 |
| O | 8.68 % | 2.44 | 4.15 | 16.03 |
| HTA | 11.41 % | 2.95 | 3.28 | 25.85 |
| LTA | 15.28 % | 4.04 | 3.82 | 31.70 |

Abbreviations other than standard chemical symbols: organic sulfur (Org. S), pyritic sulfur (Pyr. S), sulfate sulfur (Sul. S), total sulfur (Tot. S), sulfur by X-ray fluorescence (SCRF), air-dry loss (ADL), moisture (Mois.), volatile matter (Vol.), fixed carbon (Fix. C), high-temperature ash (HTA), low-temperature ash (LTA).

from the older to the younger coals in the Illinois Basin.

3. The B concentration in the coals of the Illinois Basin also increases from the older to the younger coals. This suggests that the Basin was becoming more marine (increasing in salinity) during the period of time between deposition of the older and younger coals.

Further interpretations of some of the relationships which have been noted and further elucidation of the geological parameters that have influenced the chemical characteristics of the coals should result from continuing analyses of the data. These analyses will include areal mapping of concentrations of elements and also the mapping of distributions of the elements within a single coal seam.

The average concentration of an element in the earth's crust is its clarke value. The mean value for each trace element was compared with the clarke for that element. Only three elements were enriched by at least one order of magnitude (present in an amount greater than 10 times the clarke), and three were depleted by at least one order of magnitude (present in an amount less than one-tenth the clarke). Because of the large number of variables used in calculating various clarke values, differences of less than one order of magnitude were not considered significant. The three elements enriched in coals are Cd, Se, and B. Boron is concentrated only in the coals of the Illinois Basin and probably represents a relatively high salinity of the waters in the coal swamp or of the waters which flooded the coal swamp. The only three elements found to be depleted in coals are Mn, F, and P.

An analysis of the data from the laboratory-prepared (washed) coals has allowed the grouping of the determined elements on the basis of their tendency to be concentrated with the cleanest coal or with the mineral matter. Those elements which are most closely associated with the clean coal are those with the highest "organic afinity" and include Ge, Be, and B. At the other end of the scale are those elements combined primarily in the mineral matter. This group has the least organic affinity and includes Hg, Zr, Zn, Cd, As, Pb, Mn, and Mo. The remaining elements, which are apparently associated to varying degrees with both the organic and inorganic fractions of the coal, can be divided into two groups: those which are more nearly allied with the elements with organic affinities (P, Ga, Ti, Sb, and V) and those that generally are more closely associated with the inorganic fraction (Co, Ni, Cr, Se, and Cu).

Many minerals, including aluminosilicates, carbonates, and sulfides, have been reported from coals, and the mode of occurrence of the elements that compose those minerals is evident. However, concentrations of several additional elements were noted in some of the coals studied, and the possibility that these too may be in discrete mineral phases was investigated.

Sphalerite (ZnS) has been identified as the host phase for both Zn and Cd in a number of coals, including all those which were found to contain relatively high concentrations of Zn (greater than 500 ppm). The phosphate mineral apatite, more precisely a carbonate fluorapatite, was identified in the low-temperature ashes of both the coal sample which was found to contain the largest concentration of phosphorus and the coal sample which contained the largest concentration of fluorine.

A separate phase, identified as galena (PbS), has been confirmed in the low-temperature ash of a sample high in Pb. Both Ni and Cu have been identified in intimate association (probably in solid solution) with pyrite ($FeS_2$) in several samples.

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# PRELIMINARY CHEMICAL ANALYSIS OF AQUEOUS WASTES FROM COAL CONVERSION PLANTS (A RECOMMENDED APPROACH)

**William T. Donaldson***

*Abstract*

*Limited data on aqueous wastes from coal conversion plants indicate the need for using those analytical techniques that have proven to be useful with other types of effluents. Solvent extraction-gas chromatography-mass spectrometry, with computerized data analysis, is recommended for characterization of organic constituents. Multielement techniques, such as spark source mass spectrometry or X-ray fluorescence, are recommended for elemental analyses. Such an approach, when applied to the characterization of wastes from petrochemical plants and Kraft pulp mills, showed the composition of the effluents to be significantly different from that predicted from a knowledge of raw material and manufactured products.*

## INTRODUCTION

A recent Bureau of Mines Technical Progress Report (ref. 1) states that extensive research is needed to characterize water effluents from the Synthane process (coal to gas) and to determine appropriate treatment processes. A large number of elements and organic compounds are contained in the wastewater, and the process uses a large volume of water—about 3/4 ton of water for each ton of coal processed, according to a spokesman for the Bureau of Mines. Although the proposed effluent guidelines published by the Environmental Protection Agency do not specify criteria for many elements and organic compounds, the guidelines may be more demanding in the future. Therefore, a comprehensive knowledge of effluent water composition is essential.

Even if environmental considerations were not critical, economic considerations will probably dictate that effluent water be recycled to increase efficiency. A knowledge of the water's composition, and how it changes as it is used and reused, can provide valuable insight into the changes that take place within coal conversion processes.

During the past few years, the qualitative organic analysis and multielement analysis of industrial aqueous effluents have been studied at EPA's Southeast Environmental Research Laboratory (SERL) (refs. 2, 3). Although the more traditional Chemical analyses, such as those listed in EPA's 11 Methods for Chemical Analysis of Water and Wastes (ref. 4) must be used to obtain specific measurements, we believe that the orgnic qualitative and multielement methods applied at SERL will be applicable to the comprehensive characterization of coal conversion plant aqueous wastes.

## ORGANIC ANALYSIS

Gas chromatography-mass spectrometry (GC-MS) is the best single technique for identifying moderately volatile organic compounds. To make the required investment in a mass spectrometer economically feasible, a large number of samples must be analyzed per unit time. The resulting extensive data reduction and analysis must therefore be accomplished by computers. At SERL a gas chromatograph is interfaced to a Finnigan model 1015 quadrupole mass spectrometer with a System Industries dedicated data system. A second computer is used for data analysis by comparing the mass spectra obtained from sample compounds with spectra of 8,000 identified compounds in the computer library.

A typical procedure for identifying organic compounds in wastewater involves extraction with a nonpolar solvent such as chloroform or hexane. Extracts are concentrated by evaporation to about a thousandth of their original volume, and then 1 to 10 microliters of the concentrate is injected into the GC-MS. The computer directs the mass spectrometer to scan the GC effluent every 3 seconds and stores the total ion current data ( a signal related to quantity of material in each GC peak) and corresponding mass spectra on magnetic tape. At the end of the GC-MS run, the computer retrieves the ion current information and plots it as a reconstructed gas chromatogram (RGC). A typical RGC is shown in the upper trace of figure 1. The numbers on the abscissa correspond to the mass spectra stored on tape, and the ordinate reflects the total ion monitor response.

After examining the RGC, the analyst can request

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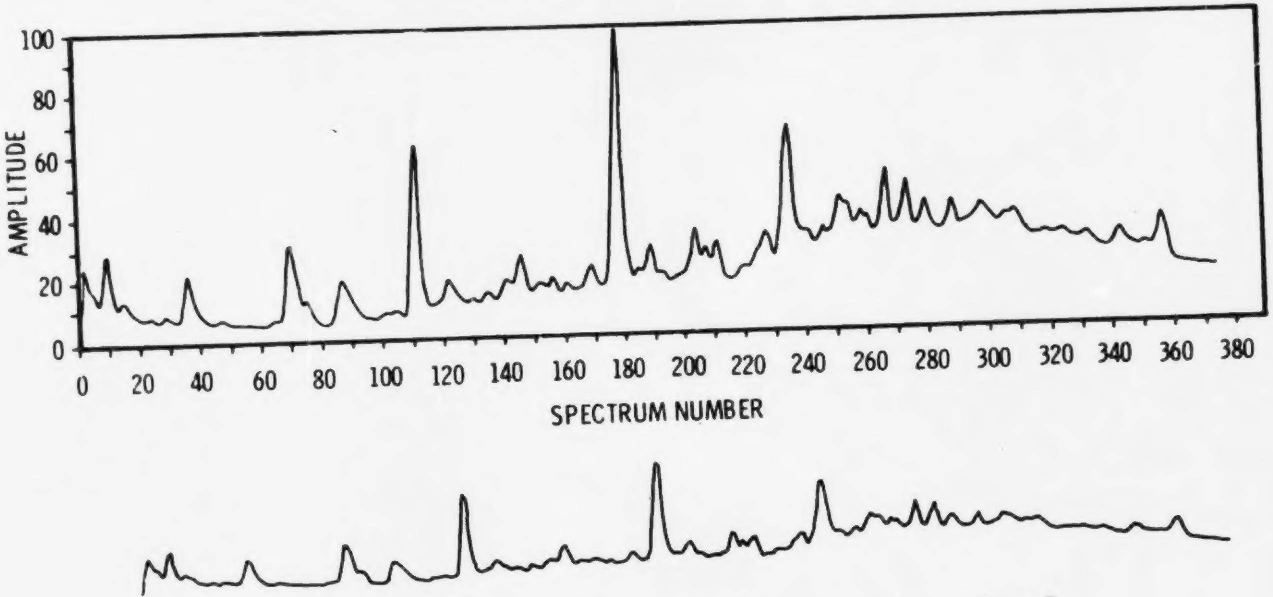
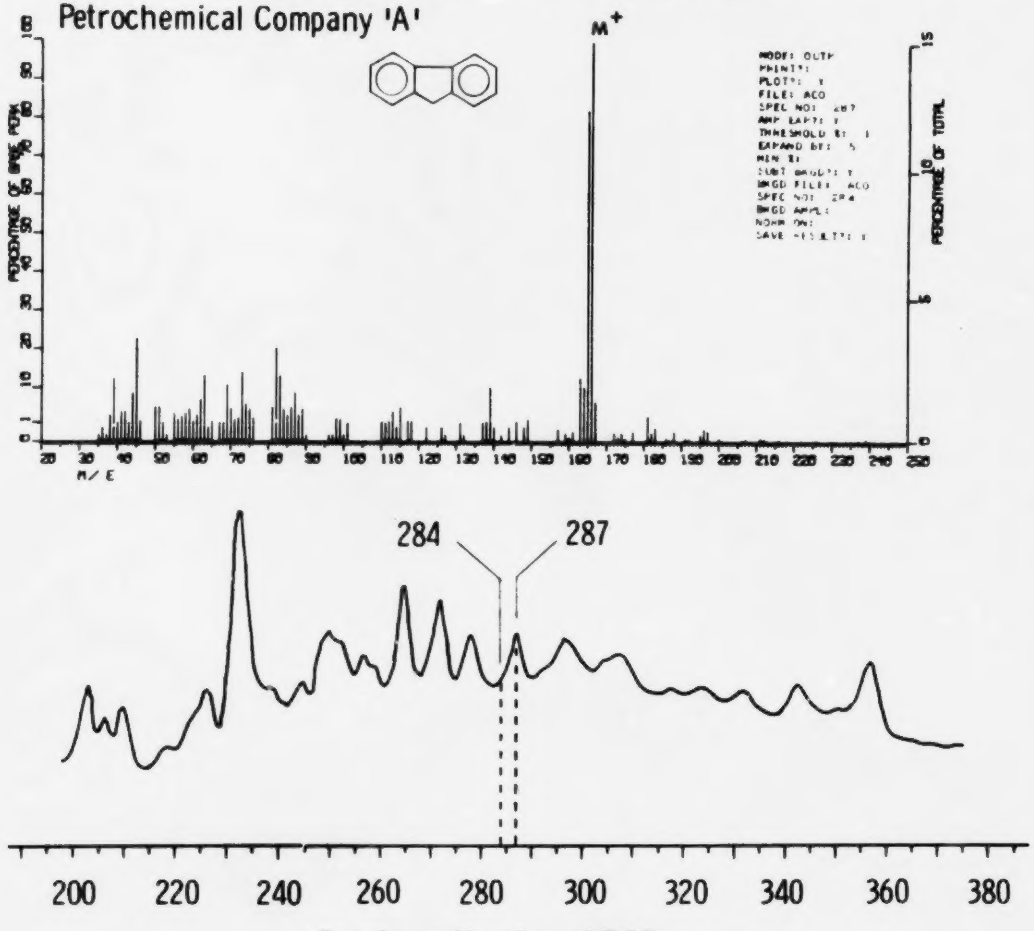


Computer-Reconstructed Gas Chromatogram [RGC]

Petrochemical Company 'A'

Real Time Ion Current Summation [ICS]

Figure 1



Mass Spectrum 287 Minus 284

Petrochemical Company 'A'

```
MODE: OUTP
PRINT?:
PLOT?:  Y
FILE: ACO
SPEC NO:  287
AMP EXP?: Y
THRESHOLD %:  1
EXPAND BY:  5
MIN %:
SUBT BKGD?: Y
BKGD FILE:  ACO
SPEC NO:  284
BKGD AMPL:
NORM ON:
SAVE RESULT?: Y
```

Spectrum Number of RGC

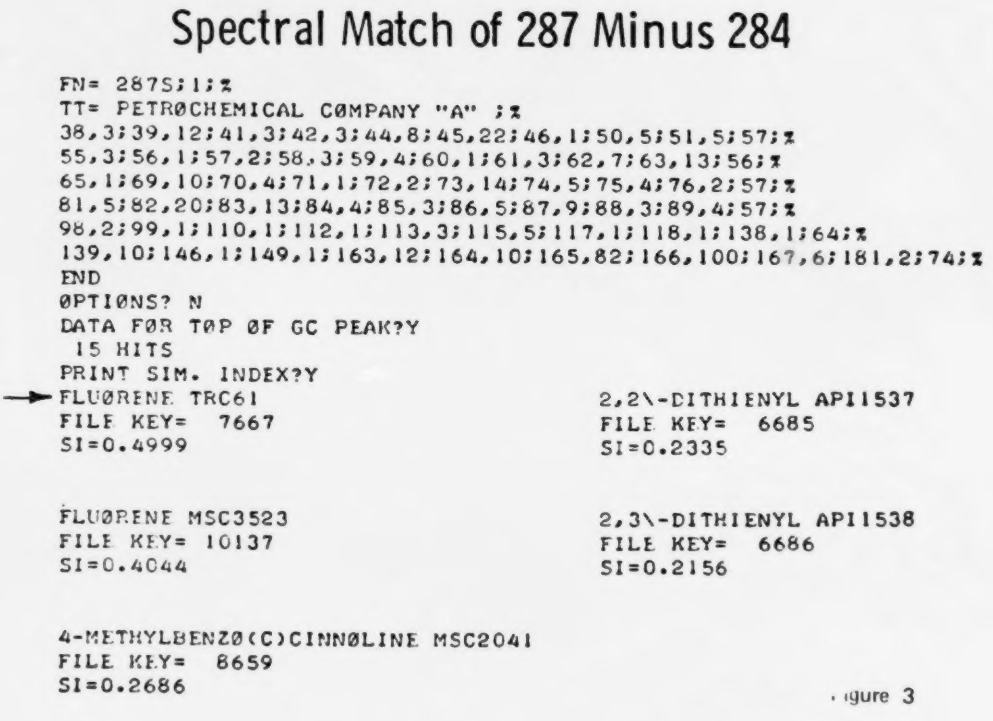
Figure 2

the computer to plot any mass spectrum by spectrum number. The spectrum numbered 287 is shown in figure 2. Contributions from impurities which were not separated by the gas chromatograph were eliminated from this spectrum by the computer by subtracting spectrum 284 taken in the GC "valley." Spectrum 287 contains quite a few discernible peaks, which have dimensions of both mass (mass to charge ratio) and amplitude (expressed in percentage of the amplitude for the largest peak). With this information, a computer should be able to select those spectra from its library that are most similar to that of the unknown compound.

The printout of the computer search for spectrum 287 is shown in figure 3. The first paragraph presents the digital representation of the unknown compound's spectrum in terms of the relative amplitude for each discernible mass. The following paragraphs list the compounds whose spectra in the computer library are most similar to that of the unknown compound. The last line of each of these paragraphs provides a similarity index (SI), which aids the analyst in selecting the most probable identification. Compounds are listed in order of their similarity indices. Note that the similarity indices for two fluorene spectra are significantly higher than that of the compound with the next highest similarity index.

Thirty-seven compounds were identified by the computer in this sample (table 1). However, the computer identification cannot be accepted as an absolute confirmation of identity. After the computer "suggests" the identity for a compound, the analyst must confirm it. He does this by first examining the spectrum to see if its fragmentation pattern is reasonable for the suggested compound. He also compares GC retention times for the unknown and suggested compounds. Finally, he can obtain other types of spectra for the compound, if it is present in high enough concentration. Chemical ionization mass spectra are usually significantly different from conventional (electron impact ionization) spectra. Infrared (IR) spectra are highly definitive, and recent advances in Fourier transform GC-IR techniques have brought its sensitivity to within one or two orders of magnitude of that of GC-MC (ref. 5).

If specific compounds or classes of compounds are of particular interest, specialized techniques of data reduction may be used to detect these materials in complex mixtures in which they may not be detectable by conventional analysis. The most



Spectral Match of 287 Minus 284

```
FN= 287S;1;%
TT= PETRØCHEMICAL CØMPANY "A" ;%
38,3;39,12;41,3;42,3;44,8;45,22;46,1;50,5;51,5;57;%
55,3;56,1;57,2;58.3;59,4;60,1;61,3;62,7;63,13;56;%
65,1;69,10;70,4;71,1;72,2;73,14;74,5;75,4;76,2;57;%
81,5;82,20;83,13;84,4;85,3;86,5;87,9;88,3;89,4;57;%
98,2;99,1;110,1;112,1;113,3;115,5;117,1;118,1;138,1;64;%
139,10;146,1;149,1;163,12;164,10;165,82;166,100;167,6;181,2;74;%
END
ØPTIØNS? N
DATA FØR TØP ØF GC PEAK?Y
 15 HITS
PRINT SIM. INDEX?Y
FLUØRENE TRC61                          2,2\-DITHIENYL API1537
FILE KEY=  7667                         FILE KEY=  6685
SI=0.4999                               SI=0.2335


FLUØRENE MSC3523                        2,3\-DITHIENYL API1538
FILE KEY= 10137                         FILE KEY=  6686
SI=0.4044                               SI=0.2156


4-METHYLBENZØ(C)CINNØLINE MSC2041
FILE KEY=  8659
SI=0.2686
```

Figure 3

common such technique is the generation of a limited mass reconstructed gas chromatogram (LMRGC). For example, in figure 4, the RGC shows peaks for compounds whose spectra contain significant numbers of any ion fragments of mass-to-charge ratios 35 to 250. Above the RGC is the LMRGC in which the computer was instructed to respond only to those spectra that contain a compound fragment with mass-to-charge ratio 149. This fragment is highly characteristic of phthalate esters. The LMRGC indicates that the sample may contain two phthalates.

The GC-MS technique is highly sensitive; acceptable spectra can be obtained for most compounds present in water at concentrations above 0.1 μg/liter by the conventional mode of operation. Unfortunately, not all compounds are amenable to solvent extraction, and some that are extracted cannot be chromatographed. Other techniques are being investigated for analysis of polar and nonvolatile compounds, but these techniques are not sufficiently developed for broad application.

## MULTIELEMENT ANALYSIS

As more consideration is given to the environmental impact of low concentrations of chemical elements, a large number of the 92 naturally occurring elements are recognized as being pollutants in certain systems. It is impossible to predict with confidence which of these elements may be present in industrial waste effluents. Analyzing for each element separately is extremely time consuming and costly. Therefore a simultaneous, multielement analytical technique is needed.

At SERL we are evaluation the spark source mass spectrometer's use in combined qualitative-quantitative analysis of water. Theoretically all chemical, nongaseous elements in any matrix can be identified and measured with the spark source mass spectrometer, provided organic materials in the sample have been reduced to their elemental components (other than carbon, hydrogen, and oxygen).

The electrical detection system of the AEI MS-702 mass spectrometer is interfaced with a DEC PDP-8/E computer. The computer program converts the electronic signal from the detector into a typed listing of all elements present and their concentrations in micrograms per liter.

Table 1

Compounds Identified in Wastewater of Petrochemical Company

| RGC Spectrum # (from Figure 1) | Compound Name |
|---|---|
| 2 | m-xylene* |
| 4 | p-xylene* |
| 10 | 1,5-cyclooctadiene |
| 16 | o-xylene* |
| 29 | isopropylbenzene (cumene) |
| 36 | styrene* |
| 47 | o-ethyltoluene |
| 65 | o-methylstyrene* |
| 70 | diacetone alcohol |
| 75 | indan* |
| 86 | 2-butoxyethanol |
| 89 | β-methylstyrene |
| 109 | indene* |
| 121 | dimethylfuran isomer |
| 129 | n-pentadecane |
| 140 | 1-methylindene* |
| 145 | 3-methylindene |
| 156 | acetophenone |
| 160 | n-hexadecane |
| 168 | α-terpineol |
| 177 | naphthalene* |
| 193 | α-methylbenzyl alcohol |
| 202 | 2-methylnaphthalene* |
| 206 | benzyl alcohol |
| 210 | 1-methylnaphthalene* |
| 221 | ethylnaphthalene isomer |
| 233 | 2,6-dimethylnapnthalene* |
| 233 | phenol* |
| 244 | methyl ethyl naphthalene isom |
| 249 | cresol isomer |
| 256 | acenaphthene |
| 266 | acenaphthalene |
| 278 | methylbiphenyl isomer |
| 287 | fluorene |
| 292 | phthalate diester |
| 296 | 3,3-diphenylpropanol |
| 356 | phthalate diester |

*Identification was confirmed with a standard.

In machine computation of data, the reliability of the mathematical constants in the computer must be considered. Before we could rely on the computer's data interpretation, we had to determine whether the sensitivity coefficients (the terms that relate electronic signal to numbers of atoms or elemental weights) were applicable over a wide range of elemental ratios. Two solutions were prepared containing 16 elements in equal concentrations in one solution and 18 different elements in equal concentrations in the other solution. The solutions were diluted and mixed into two samples, one solution containing 100 parts of the 16 elements in group I to 1 part of the 18 elements in group II, and one solution containing 100 parts of elements in group II to 1 part of elements in group I. The ratio of concentrations of group I elements to those of group II elements therefore differed by a factor of 10,000 between the two solutions.

When the two samples were analyzed by the spark source mass spectrometer, using the sensitivity coefficients established for solutions containing single elements, no serious matrix effects were indicated. The data are displayed in figures 5 and 6. Each point represents the average of 9 determinations.

Figure 7 represents the computer printout for one of the analyses performed on sample No. 1. Note that for confirmation the computer considers

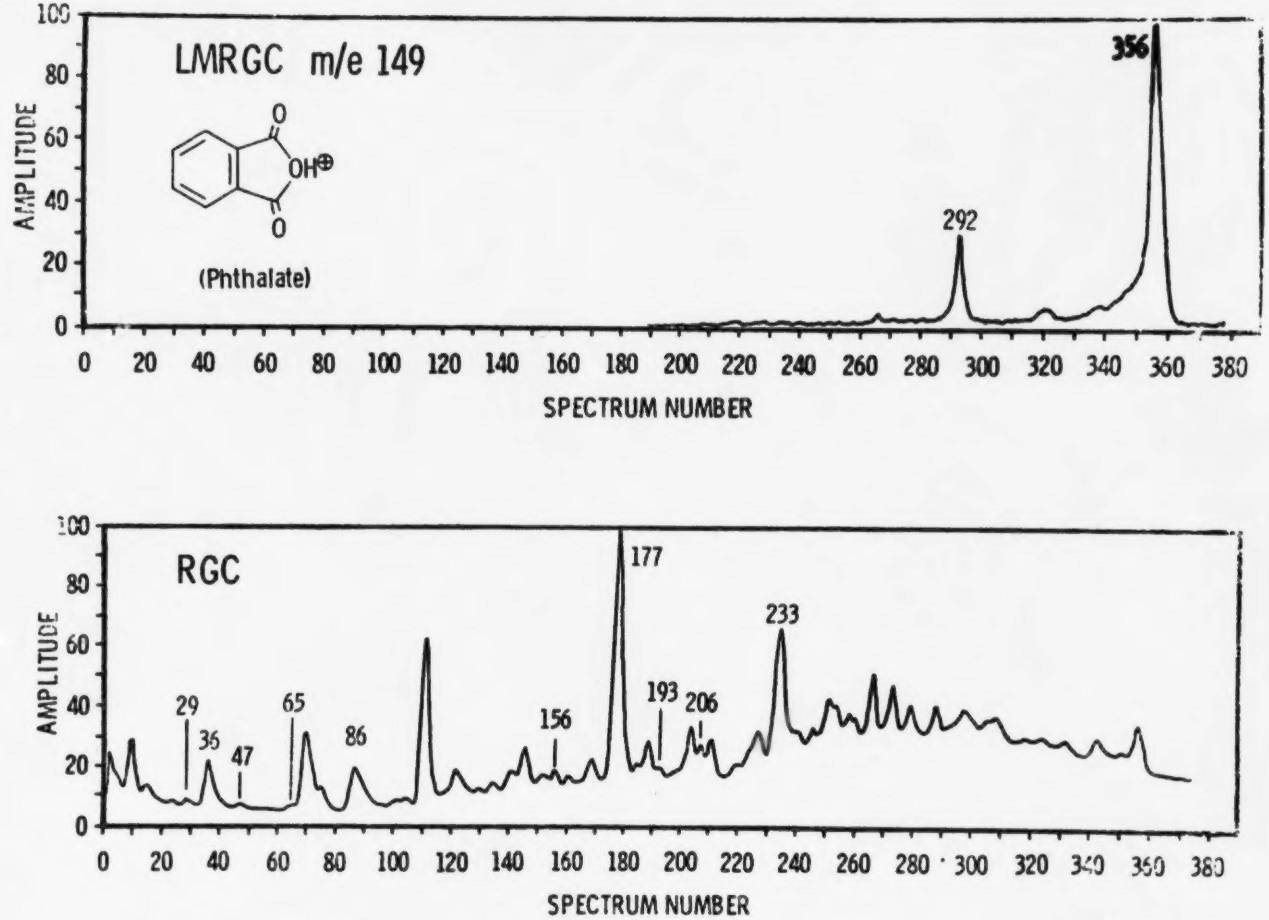
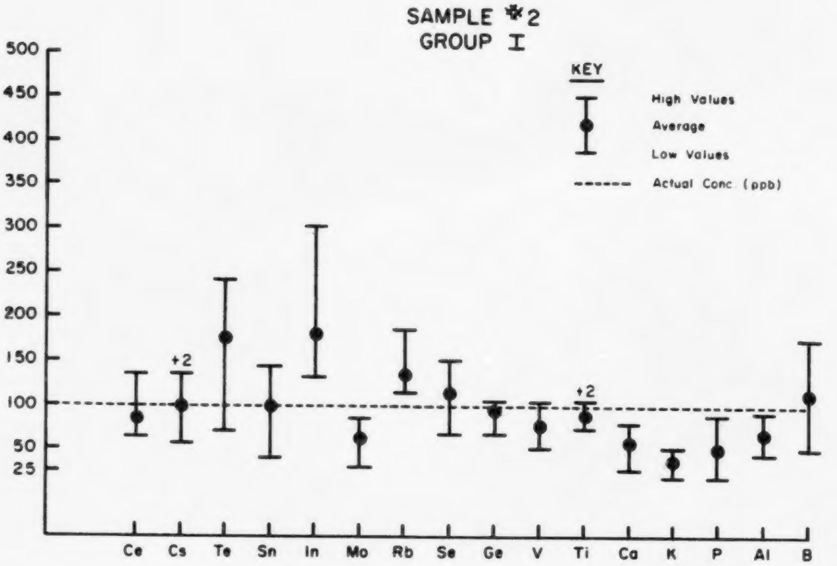
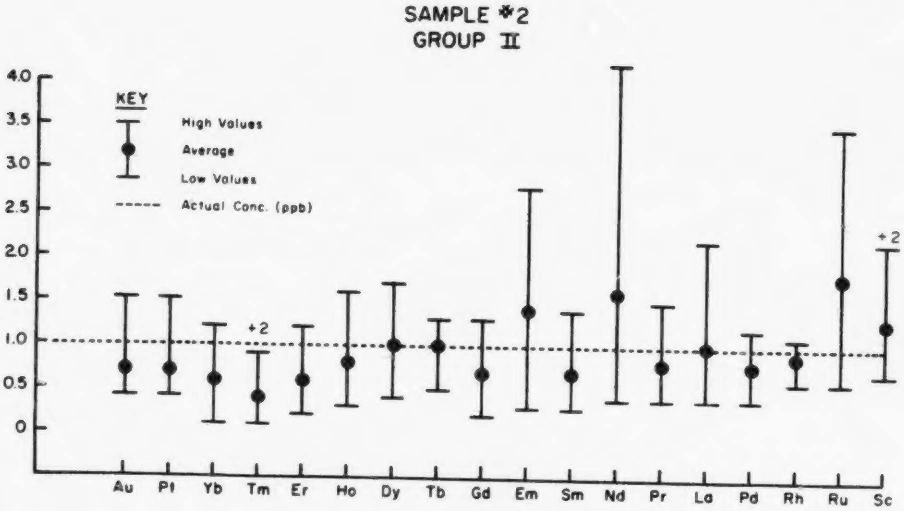

LMRGC m/e 149
OH⊕
O
O
(Phthalate)
356
292
AMPLITUDE
0
20
40
60
80
100
SPECTRUM NUMBER
0 20 40 60 80 100 120 140 160 180 200 220 240 260 280 300 320 340 360 380
RGC
177
233
29
36
47
65
86
156
193
206
AMPLITUDE
0
20
40
60
80
100
SPECTRUM NUMBER
0 20 40 60 80 100 120 140 160 180 200 220 240 260 280 300 320 340 360 380

Figure 4



SAMPLE #2
GROUP I



SAMPLE #2
GROUP II

Figure 5

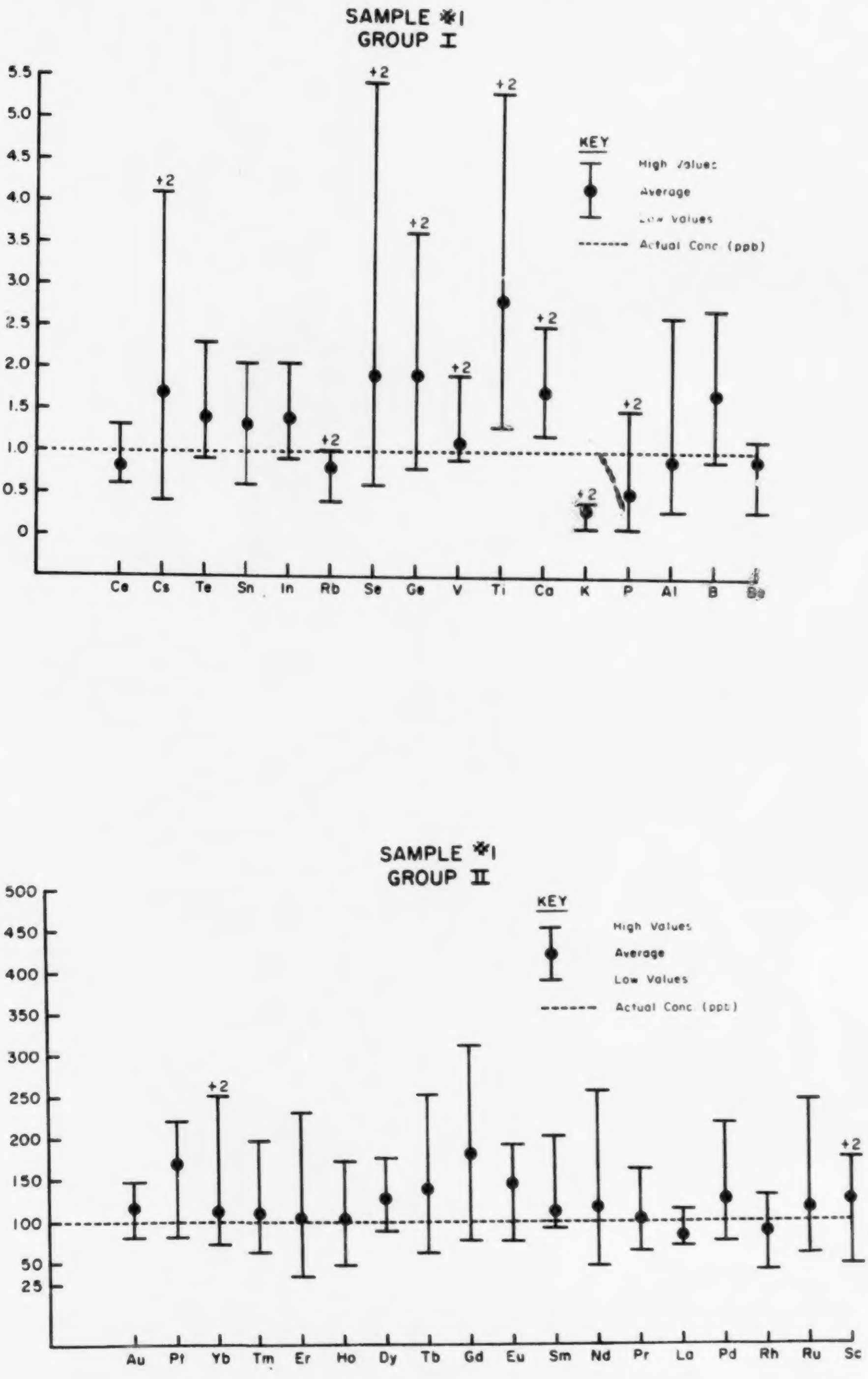

SAMPLE #1
GROUP I
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0
+2
KEY
High Values
Average
Low Values
Actual Conc (ppb)
Ce
Cs
Te
Sn
In
Rb
Se
Ge
V
Ti
Ca
K
P
Al
B
Be
SAMPLE #1
GROUP II
500
450
400
350
300
250
200
150
100
50
25
+2
KEY
High Values
Average
Low Values
Actual Conc (ppt)
Au
Pt
Yb
Tm
Er
Ho
Dy
Tb
Gd
Eu
Sm
Nd
Pr
La
Pd
Rh
Ru
Sc

Figure 6

Modified Interpretation of +1 and +2 Ion Data

```
JULY                  CONCEN       ++?   CONFIRM     CHECK     COMPLEX
SEVENTEEN            TRATIONS     +++?  ISOTOPES    OVERLAP     IONS


 197  GOLD              88.75      2+       -       -----NO MATRIX-----
 195  PLATINUM         258.88      2+      YES
 191  IRIDIUM            2.88      NO      NO
 189  OSMIUM             8.15      NO      NO
 182  TUNGSTEN           7.29      NO      NO
 175  LUTECIUM            .52      NO      NO
 172  YTTERBIUM        146.22      2+      YES
  85  THULIUM           87.23      2+       -        169(  85)
 167  ERBIUM           136.39      2+      YES
 165  HOLMIUM          102.67      2+       -
 163  DYSPROSIUM       230.85      2+      YES
 159  TERBIUM          126.73      2+       -
 158  GADOLINIUM       222.27      2+      YES
 153  EUROPIUM         104.12      2+      YES
 147  SAMARIUM         116.74      2+      NO
 143  NEODYMIUM        152.42      2+      NO
 141  PRASEODYME        96.06      2+       -
 140  CERIUM             2.04      NO      NO
 139  LANTHANUM         67.01      2+       -
 138  BARIUM              .31      NO      NO
  66  CESIUM              .49      2+       -
 128  TELLURIUM          2.81      NO      YES
 118  TIN                1.89      NO      NO
 115  INDIUM             2.32      NO      NO
 111  CADMIUM            3.80      NO      NO
 105  PALLADIUM        101.65      2+      NO
  51  RHODIUM           52.31      2+       -        103(  51)
  50  RUTHENIUM         77.83      2+      NO        101(  50)
  95  MOLYBDENUM         1.41      NO       -        190(  95)
  89  YTTRIUM      STANDARD
  43  RUBIDIUM            .66      2+       -         85(  43)
                                                     170(  85)
  28  MANGANESE           .01      2+       -         55(  28)
                                                      82(  28)
  25  VANADIUM            .70      2+       -         51(  25)
                                                      76(  25)
  24  TITANIUM           1.47      2+      NO         47(  24)
                                                      71(  24)
  23  SCANDIUM         103.97      2+       -         45(  23)
  20  CALCIUM            3.86      NO      NO         40(  20)
                                                      60(  20)
  20  POTASSIUM           .21      2+      YES        39(  20)
                                                      82(  41)
  18  CHLORINE         220.39      2+       -         35(  18)
  16  PHOSPHORUS          .41      2+       -         31(  16)
  14  ALUMINIUM           .58      2+       -         27(  14)
  12  SODIUM              .63      2+       -         23(  12)
  19  FLUORINE            .03      NO       -         38(  19)
                                                      57(  19)
  16  OXYGEN            19.84      NO       -         34(  17)
                                                      51(  17)

  14  NITROGEN          27.55      NO       -         28(  14)
                                                      30(  15)
                                                      45(  15)
  11  BORON              1.18      NO      NO         20(  10)
                                                      30(  10)
   9  BERYLLIUM           .94      NO       -

                        END OF RUN
```

Figure 7

Table 2

Mass Spectrometric Analyses of the Benzene-Soluble Tar, Volume-Percent

| Structural Type (Includes Alkyl Derivatives) | Run HP-1 No. 92, Illinois[a] No. 6 coal | Run HPL No. 94, Lignite | Run HPM No. 111, Montana Subbituminous Coal | Run HP-118 No. 118,[a] Pittsburgh Seam Coal |
|---|---|---|---|---|
| Benzenes | 2.1 | 4.1 | 3.9 | 1.9 |
| Indenes | [b]8.6 | 1.5 | 2.6 | [b]6.1 |
| Indans | 1.9 | 3.5 | 4.9 | 2.1 |
| Naphthalenes | 11.6 | 19.0 | 15.3 | 16.5 |
| Fluorenes | 9.6 | 7.2 | 9.7 | 10.7 |
| Acenaphthenes | 13.5 | 12.0 | 11.1 | 15.8 |
| 3-ring aromatics | 13.8 | 10.5 | 9.0 | 14.8 |
| Phenylnaphthalenes | 9.8 | 3.5 | 6.4 | 7.6 |
| 4-ring pericondensed | 7.2 | 3.5 | 4.9 | 7.6 |
| 4-ring catacondensed | 4.0 | 1.4 | 3.0 | 4.1 |
| Phenols | 2.8 | 13.7 | 5.5 | 3.0 |
| Naphthols | (b) | 9.7 | 9.6 | (b) |
| Indanols | .9 | 1.7 | 1.5 | .7 |
| Acenaphthenols | - | 2.5 | 4.6 | 2.0 |
| Phenanthrols | 2.7 | - | .9 | - |
| Dibenzofurans | 6.3 | 5.2 | 5.6 | 4.7 |
| Dibenzothiophenes | 3.5 | 1.0 | 1.5 | 2.4 |
| Benzonaphthothiophenes | 1.7 | - | - | - |
| N-heterocyclics[c] | (10.8) | (3.8) | (5.3) | (8.8) |
| Average molecular weight | 212 | 173 | 230 | 202 |

[a] Spectra indicate traces of 5-ring aromatics.

[b] Includes any naphthol present (not resolved in these spectra).

[c] Data on N-free basis since isotope corrections were estimated.

doubly-charged and triply-charged ions as well as isotopes of the element other than the one used for the computation of concentration.

## PRELIMINARY APPLICATION TO COAL GASIFICATION EFFLUENTS

In analyzing waste products and effluents from the Synthane Process, the Bureau of Mines chemists used organic mass spectrometry for analysis of tars, and spark source mass spectrometric analyses were performed on aqueous wastes at the SERL (ref. 1). Table 2 shows organic compounds found in the tars. These compounds must also be considered in wastewater analyses. Table 3 lists the 20 chemical elements found to be present in concentrations greater than 1 part per billion in the aqueous effluent.

Until the composition and variation in composition of aqueous wastes from coal gasification are firmly established, comprehensive analyses such as these are recommended. After knowledge of the variation in composition is known, then an appropriate monitoring program can be set up and comprehensive analyses will be required only where experience dictates.

If such an approach is followed, the coal gasification industry will be a leader in providing information to assess the environmental impact of aqueous effluents. It will also be in a good position to increase cost-effectiveness of resource management while protecting the aquatic environment.

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Table 3

Trace Elements in Condensate from
an Illinois No. 6 Coal Gasification Test

| | No. 1 | No. 2 | Average (by weight) |
|---|---|---|---|
| Ppm: | | | |
| Calcium | 4.4 | 3.6 | 4 |
| Iron | 2.6 | 2.9 | 3 |
| Magnesium | 1.5 | 1.8 | 2 |
| Aluminum | 0.8 | 0.7 | 0.8 |
| Ppb: | | | |
| Selenium | 401 | 323 | 360 |
| Potassium | 117 | 204 | 160 |
| Barium | 109 | 155 | 130 |
| Phosphorus | 82 | 92 | 90 |
| Zinc | 44 | 83 | 60 |
| Manganese | 36 | 38 | 40 |
| Germanium | 32 | 61 | 40 |
| Arsenic | 44 | 28 | 30 |
| Nickel | 23 | 34 | 30 |
| Strontium | 33 | 24 | 30 |
| Tin | 25 | 26 | 20 |
| Copper | 16 | 20 | 20 |
| Columbium | 7 | 5 | 6 |
| Chromium | 4 | 8 | 6 |
| Vanadium | 4 | 2 | 3 |
| Cobalt | 1 | 2 | 2 |

Protection Agency Research Report EPA-R2-73-277, August 1973.

3. C. E. Taylor and W. J. Taylor, "Multielement Analysis of Environmental Samples by Spark Source Mass Spectrometry," Environmental Protection Agency Research Report EPA 660/2-74-002, January 1974.
4. "Methods for Chemical Analysis of Water and Wastes," Environmental Protection Agency Manual, Analytical Quality Control Laboratory, Cincinnati, Ohio, July 1971.
5. L. V. Azarraga and A. C. Mc Call, "Infrared Transform Spectrometry of Gas Chromatography Effluents," Environmental Protection Agency Research Report EPA 660/2-73-034, January 1974.

14 May 1974

## Session III:

## ENVIRONMENTAL ASPECTS OF SPECIFIC FUEL CONVERSION SYSTEMS

David H. Archer, Ph.D.
Session Chairman

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# SOME IMPLICATIONS OF ENVIRONMENTAL REGULATORY ACTIVITIES ON COAL CONVERSION PROCESSES

E. S. Rubin and F. C. McMichael[*]

*Abstract*

*Existing U.S. environmental regulatory policies for air and water pollution control are reviewed in detail from the point of view of their potential implications on coal conversion processes presently under development in this country. The discussions focus on standards of environmental quality, new source performance or discharge standards, and the interactive roles of Federal, State, and local authorities in defining and implementing environmental control programs. Elements of existing regulatory activities are shown to impose potentially conflicting or inconsistent requirements that may be counterproductive to environmental quality. Several areas for further study are indicated.*

## INTRODUCTION

### *Background and Scope of Discussion*

Coal conversion plants are soon to be part of the fuel technology of the United States. They are envisioned to be a complex of physical-chemical unit operations that offer the promise of clean fuels to our society. However, it is important that the environmental impact of coal as a fuel is not simply transferred from existing industries to new kinds of facilities at other locations.

Table 1 lists several of the more well-known processes that are under development in the United States to produce clean fuels from coal, including high-Btu gas, low-Btu gas, and synthetic liquids. While it is clear that commercial production and utilization of low-sulfur, low-ash synthetic fuels will significantly help enhance environmental quality at many sites, it is also clear that there are potential environmental problems associated with the conversion processes themselves. These potential problems are quite sizeable and must be carefully evaluated (refs. 1-4). Air pollution, water pollution, solid waste disposal, and thermal pollution problems, already familiar to much of today's in-place industrial technology, will be among the problems also encountered in coal conversion processing.

Environmental control measures in these areas will take place within a regulatory framework established by Federal, State, and local agencies charged with defining and implementing environmental control policy with respect to one or more areas of impact. To gain some perspective on the possible shape of such a policy, this paper is directed at a review of some of the recent environmental regulatory activities that are likely to affect new coal conversion processes, including an examination of some of the implications of these activities on coal conversion process requirements and resulting environmental impacts. Discussions will be restricted principally to a consideration of problems in air and water pollution, in that these are the areas in which regulatory activities are at present most strongly focused. These two problems are strongly related to one another, and it is the nature of this relationship and the ability of regulatory policy to deal with it (as with the relationship to other problems such as solid waste disposal) that will be held up for particular attention.

### *Nature and Sources of Pollutants from Coal Conversion Processes*

Tables 2 and 3 indicate the major air and water pollutants known or suspected to be associated with coal gasification or liquefaction processes (refs. 4-9). Table 2 for air pollutants indicates whether release of the pollutant occurs principally as a result of the coal conversion process or as a result of fuel combustion. The latter includes auxiliary combustion required for the conversion process (e.g., to provide utilities such as steam and electric power), as well as end-use combustion of the synthetic fuel products. Since boilerhouse steam, raised by direct combustion, is required to operate pollution control systems on plant process streams, it is appropriate to view combustion-generated pollutants as resulting in part from the need for other environmental control measures. In this light, tradeoffs among different pollutants and environmental media become somewhat more apparent, as will be further illustrated later. First, the following paragraphs briefly review the major sources of air and water pollutants in coal conversion processing.

#### *Air Pollutants*

*Sulfur dioxide* is emitted principally from the

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Table 1. Selected Coal Conversion Processes*

| NAME | DEVELOPER/SPONSOR | STATUS |
|---|---|---|
| HIGH-BTU GASIFICATION | | |
| BI-GAS | BCR/OCR-AGA | Pilot (u/c) |
| COGAS | FMC Corp. | Pilot |
| $CO_2$ Acceptor | Consol/OCR-AGA | Pilot |
| HYGAS | IGT/OCR-AGA | Pilot |
| Lurgi | Lurgi | Comm'l (prop.) |
| Molten Salt | M.W. Kellogg | Bench |
| Synthane | BuMines | Pilot (u/c) |
| LOW-BTU GASIFICATION | | |
| Agglomerating Gasifier | Westing./OCR-Private | Pilot (u/c) |
| Atgas | ATI/EPA | Bench |
| Entrained Gasifier | CE/OCR | Design |
| Koppers-Totzek | Koppers | Comm'l |
| Lurgi | Lurgi | Comm'l |
| Stirred Fixed Bed | BuMines | Bench |
| LIQUEFACTION | | |
| COED | FMC/OCR | Pilot |
| Direct Hydrogenation | Univ Utah/OCR | Bench |
| Hydrogen Donor | Exxon | Bench |
| Project Gasoline | Conoco | Pilot |
| Solvent Refined Coal | P&M/So. Svcs/OCR | Pilot |
| Turbulent Catalytic | BuMines | Bench |

*Ref. 2

tailgas stream of the sulfur recovery plant and from stack gases of auxillary systems requiring fuel oxidation. The latter most prominantly includes coal pretreatment processes, plant boilerhouse, and miscellaneous process heaters and burners fired with sulfur-bearing fuels.

*Particulate matter* can be released to the atmosphere both as a fugitive dust and as a process or combustion-based stack emission. Fugitive emissions would be most likely to occur at receiving, handling, and storage areas for coal, solid wastes, or solid products, but could also occur as leakages from process equipment trains. Process stack emissions would include the exhaust streams of pollution control equipment such as scrubbers, precipitators, and baghouses associated with unit operations including coal driers, pulverizers, gasifiers, prilling towers, and the like (details of which would vary from process to process). Again, fuel combustion would provide another potentially significant source of particulate where furnaces or boilers are fired with solid or perhaps liquid fuels.

*Nitrogen oxide* emissions result from combustion in boilers and process heaters fired on either gaseous, liquid, or solid fuels. Emissions would depend on the fuel type, boiler design, various combustion parameters, and the fuel nitrogen content.

*Hydrocarbon* emission in coal conversion could occur from liquid product storage areas, from leakage at valves, flanges, and seals of the assorted pipes, vessels, pumps, and compressors throughout the plant, and from evaporation of hydrocarbon liquids dissolved in effluent or cooling streams. Combustion-generated hydrocarbons may also emanate from furnace and boiler stacks as a result of incomplete combustion.

*Carbon monoxide*, produced in large quantities in coal gasification processes, is a prime fuel constituent, and no significant release to the atmosphere would be expected from the process stream. As with

Table 2. Types and Sources of Potential Air Pollutants from Coal Conversion Processes

| Pollutant | Process-Generated | Combustion-Generated |
|---|---|---|
| Particulate Matter | x | x |
| Sulfur Oxides | x | x |
| Reduced Sulfur Compounds | x | |
| Nitrogen Oxides | | x |
| Hydrocarbons | x | x |
| Carbon Monoxide | x | x |
| Trace Metals | x | x |
| Odors | x | |
| Other Gases (incl. $NH_3$, HCN, HCl) | x | |

Table 3 Composition of Wastewaters Representative of Coal Conversion Process Waters

| Pollutant | Ammonia Liquor* (mg/liter) | Synthane By-Product Water** (mg/liter) |
|---|---|---|
| pH | 8.3-9.1 | 7.9-9.3 |
| COD | 2,500-10,000 | 1,700-43,000 |
| Ammonia | 1,800-4,300 | 2,500-11,000 |
| Cyanide | 10-37 | 0.1-0.6 |
| Thiocyanate | 100-1,500 | 21-200 |
| Phenols | 410-2,400 | 200-6,600 |
| Sulfide | 0-50 | N/D |
| Alkalinity (as $CaCO_3$) | 1,200-2,700 | N/D |
| Specific conductance (as umho/cm) | 11,000-32,000 | N/D |

*Ref. 15
**Ref. 6
N/D = Not determined

hydrocarbons, some amount of CO may be released to the atmosphere as a result of incomplete combustion of fuel in plant heaters and boilers.

*Reduced sulfur compounds* (principally $H_2S$, COS, and $CS_2$) occur in the initial product streams of virtually all coal conversion processes, from which they are nominally stripped and converted to a recoverable substance. Potentially, emissions could occur as a result of incomplete removal (such as from a $CO_2$ exhaust steam), leakage from valves and seals, and evaporation (at cooling towers) from scrubbing water containing dissolved gases and from water quenching of solid process residues or slags containing sulfur.

*Trace element* emissions of such substances as mercury, berillium, arsenic, cadmium, vanadium, and other heavy metals (all of which are contained in coal in small amounts) is a subject about which relatively little is presently known, although some work in this area is emerging (refs. 10-13). Studies of trace metal emissions from coal-fired boilers, for example, indicated that most of the mercury and possibly other trace metals in coal are vaporized and emitted in the stack flue gas. The fate of these substances in coal conversion plants requires further detailed study.

Finally, other gaseous emissions, especially *hydrogen cyanide* and *ammonia* (as well as hydrogen chloride and gaseous odorants), may also be associated with coal conversion plants, as some developing emission regulations anticipate (ref. 9). Such emissions would again most likely occur via evaporation from the cooling of scrubbing liquors, although not much quantitative information is currently available.

### *Water Pollutants*

Wastewaters from coal conversion processes can originate from four sources: (1) moisture in the coal, (2) water of constitution, (3) water added for stoichiometric process requirements, and (4) water introduced for byproduct recovery or gas scrubbing. The first two sources alone, for example, may contribute a flow of 20 to 30 gallons per ton without considering condensates formed by other mechanisms (ref. 14). Since these process waters come into contact with contaminates in coal, they are the principal source of pollution, as opposed to nonprocess waters such as used for indirect cooling.

Since coal conversion processes, however, are net *consumers* of water, all streams could theoretically be recycled for use in the process, and no wastewater streams need emanate from the plant. Effluent streams occur in practice because it is often difficult or impossible, for reasons of technology and/or economics, to recycle all wastewaters consumptively and control all stream flows to yield the precise overall stoichiometric water requirement.

The complex composition of the process waters from coal reflects the fact that water is a universal solvent. In many ways, coal process waters have an inorganic composition as saline as sea water. They include practically all the organic compounds found in coal. Any condensate water should be expected to have a composition that falls within the range of composition shown in table 3.

A major wastewater stream from the byproduct coking industry, waste ammonia liquor, may offer some hint to the expected composition of coal conversion plant process wastewaters. The similarity in table 3 of ammonia liquor contaminants with data reported for Synthane byproduct waters further suggests the complexity of coal conversion plant wastewater.

## REGULATORY POLICIES FOR AIR AND WATER POLLUTION CONTROL

Table 4 summarizes the nature of existing regulatory policies for air and water pollution control. Generically, our present environmental policies take the form of two types of regulations: (1) standards of environmental quality, and (2) standards limiting the discharge of specified substances to the environment, either by specifying a maximum allowable discharge rate and/or concentration, or by requiring specific types of control equipment or system design.

The two types of standards may or may not be related to, or consistent with, one another. In air pollution, environmental air quality standards often do provide the philosophical and quantitative basis for the national emission control programs presently in place. In water pollution, however, present control programs are in practice based mainly on considerations of technological capability rather than on compatibility with water quality standards. The following paragraphs elaborate on the current types of environmental and discharge standards that may affect coal conversion processing.

### *Standards of Environmental Quality*

Standards of environmental quality for air and water are established by local, State, and Federal authorities. The 1970 Clean Air Act Amendments have given the Federal Government the leading role in

Table 4. Summary of Current U.S. Regulatory Policy for Air and Water Pollutants

(A User's Guide to the Regulatory Lexicon)

| ELEMENTS OF AIR POLLUTION CONTROL POLICY | ELEMENTS OF WATER POLLUTION CONTROL POLICY |
|---|---|
| AMBIENT AIR QUALITY STANDARDS (AAQS) | RECEIVING WATER QUALITY STANDARDS (RWQS) |
| Nat'l Primary Standards<br>Nat'l Secondary Standards<br>Non-degradation Standards<br>State & Local Standards | U.S. PUBLIC HEALTH SERVICE DRINKING WATER STANDARDS |
| EMISSION STANDARDS | EFFLUENT LIMITATIONS |
| New Source Performance Standards (NSPS) | New Source Performance Standards (NSPS) |
| | Best Practicable Technology<br>Best Available Technology<br>Pretreatment Standards |
| | Zero Discharge |
| Hazardous Pollutants | Toxic Substances |
| State and Local Standards | State and Local Standards |
| THRESHOLD LIMIT VALUE | |

the area of air by requiring EPA to promulgate national ambient air quality standards (AAQS), including primary standards that are protective of human health, and secondary standards protective of human welfare (table 5). Emission control strategies must be designed to achieve primary standards by mid-1975 and secondary standards as soon as possible thereafter. State and local authorities, however, may promulgate air quality standards that are more stringent than the national standards, or that apply to pollutants or averaging times not covered by Federal regulations. Table 6 illustrates this for six States located in the eastern coal fields of the United States (fig. 1).

Federal responsibility also includes insuring the "nondegradation" of air with a quality better than the national standards. In 1973, EPA proposed four alternative plans to prevent "significant deterioration" (ref. 18), although no final plan has yet been adopted. More recently, a modified version of that plan was suggested by EPA, as summarized in table 7. It has also invited Congress to reexamine this issue.

In the water pollution area, State-imposed receiving water quality standards (RWQS) also apply to all of the Nation's waterways, but do not usually serve as the basis for establishing source effluent limitations. The Federal Water Pollution Control Act Amendments of 1972 aim to "restore and maintain the chemical, physical, and biological integrity of the Nation's waters." It is the "national goal that the discharge of pollutants into the navigable waters be eliminated by 1985." However, by July 1, 1983, it is the "national goal wherever attainable to have water quality which provides for the protection and propagation of fish, shellfish, and wildlife and provides for recreation in and on the water." Furthermore, it is "the national policy that the

Table 5. National Ambient Air Quality Standards

| Contaminant | Averaging Interval | Primary Standard | | Secondary Standard | |
|---|---|---|---|---|---|
| | | $\mu g/m^3$ | ppm (by vol.) | $\mu g/m^3$ | ppm (by vol.) |
| Suspended Particulates | 1 year | 75 | -- | 60 | -- |
| | 24 hr. | 260 | -- | 150 | -- |
| Sulfur Dioxide | 1 year | 80 | 0.03 | -- | -- |
| | 24 hr. | 365 | 0.14 | -- | -- |
| | 3 hr. | -- | -- | 1,300 | 0.5 |
| Carbon Monoxide | 8 hr. | 10,000 | 9.0 | 10,000 | 9.0 |
| | 1 hr. | 40,000 | 35.0 | 40,000 | 35.0 |
| Photochemical Oxidant | 1 hr. | 160 | 0.08 | 160 | 0.08 |
| Hydrocarbons (non-methane) | 3 hr. (6-9 a.m.) | 160 | 0.24 | 160 | 0.24 |
| Nitrogen Dioxide | 1 year | 100 | 0.05 | 100 | 0.05 |

Notes: 1. All values other than annual values are maximum concentrations *not to be exceeded more than once per year*.

2. PPM values are approximate only.

3. All concentrations relate to air at standard conditions of 25°C temperature and 760 millimeters of mercury pressure.

4. Annual average refers to arithmetic mean for gases and geometric mean for particulates.

discharge of toxic pollutants in toxic amounts be prohibited." There is an attempt to separate or decouple effluent standards and receiving water quality standards. The water quality standards program has been expanded to include intrastate as well as interstate waters. The States individually set their own receiving water standards, which must be approved by the EPA, and are expected to meet the goals and deadlines of the new law. They are also to be used to measure the effectiveness of effluent limitations. The EPA and the States may prescribe more stringent effluent limitations to protect high quality bodies of water when it is evident that receiving water quality does not provide for the beneficial uses of human recreation and fish propagation.

There are also general water quality criteria designed to protect the water uses of streams. These limits typically refer to the elimination of floating solids, films, scums, bottom deposits, and objectionable odors. States also set specific limits, sometimes daily averages as well as monthly averages, for particular pollutants. Commonly, all States will set limits on pH, temperature, and dissolved oxygen. However, the levels and averaging periods are often different. Specific wastewater parameters such as phenol, ammonia, and cyanide may be set at different limits on the same river by different States, e.g., the phenol limit for West Virginia and Pennsylvania (table 8).

Special types of air and water quality standards indicated in table 4 are the U.S. Public Health Service Drinking Water Standards and Threshold Limit Value (TLV) standards for air pollutants. Public Health Service drinking standards specify the maximum concentration of various substances acceptable for drinking water use on interstate carriers. (They are also the generally accepted comparison for rating all public water supplies.) Threshold limit values define maximum concentrations of air pollutants for industrial exposure as required under the Occupational Safety and Health Act, and are usually considerably less stringent than standards applying to the population as a whole.

Table 6. Selected Local Ambient Air Quality Standards for Six Eastern States

| State | Pollutant | Standard (ug/$m^3$)[*] |
|---|---|---|
| Illinois | N/A | N/A |
| Indiana | N/A | N/A |
| Kentucky | Florides | 80 ppm (30-da avg) |
| | Hydrogen Sulfide | 0.01 ppm (1-hr avg) |
| | Hydrogen Fluoride | 0.82 (30-da)<br>1.64 (1-wk)<br>2.86 (24-hr)<br>3.68 (12-hr) |
| Ohio | N/A | N/A |
| Pennsylvania | Settled Particles | 800 (1-yr)<br>1500 (30-da) |
| | Suspended Particulates[**] | 350 (4-hr) |
| | Lead | 5 (30-da) |
| | Beryllium | 0.01 (30-da) |
| | Sulfates ($H_2SO_4$) | 10 (30-da)<br>30 (24-hr) |
| | Fluorides (as HF) | 5 (30-da) |
| | Hydrogen Sulfide | 0.005 ppm (24-hr)<br>0.1 ppm (1-hr) |
| West Virginia | N/A | N/A |

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[*] Maximum value, not to be exceeded

[**] Allegheny County only

Table 7. Proposed Nondegradation Standards[*]

| Applicability | Maximum Allowable Degradation | |
|---|---|---|
| | Sulfur Dioxide | Particulate Matter |
| Zone 1 Areas<br>(Restricted Development) | 2 ug/$m^3$(annual avg)<br>5 ug/$m^3$(24-hr avg)<br>25 ug/$m^3$(3-hr avg) | 5 ug/$m^3$ (annual avg)<br>10 ug/$m^3$(24-hr avg) |
| Zone 2 Areas<br>(Modest Development) | 15 ug/$m^3$(annual avg)<br>100 ug/$m^3$(24-hr avg)<br>300 ug/$m^3$(3-hr avg) | 10 ug/$m^3$(annual avg)<br>30 ug/$m^3$(24-hr avg) |
| Zone 3 Areas<br>(Concentrated Development) | Up to Secondary Ambient Air Quality Standards<br>(1300 ug/$m^3$, 3-hr avg) | (60 ug/$m^3$,annual avg)<br>(150 ug/$m^3$, 24-hr avg) |

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[*] Ref.19. Degradation for Zones 1 and 2 are essentially the same levels formally proposed by EPA as alternative definitions of "significant deterioration." (Ref 18).

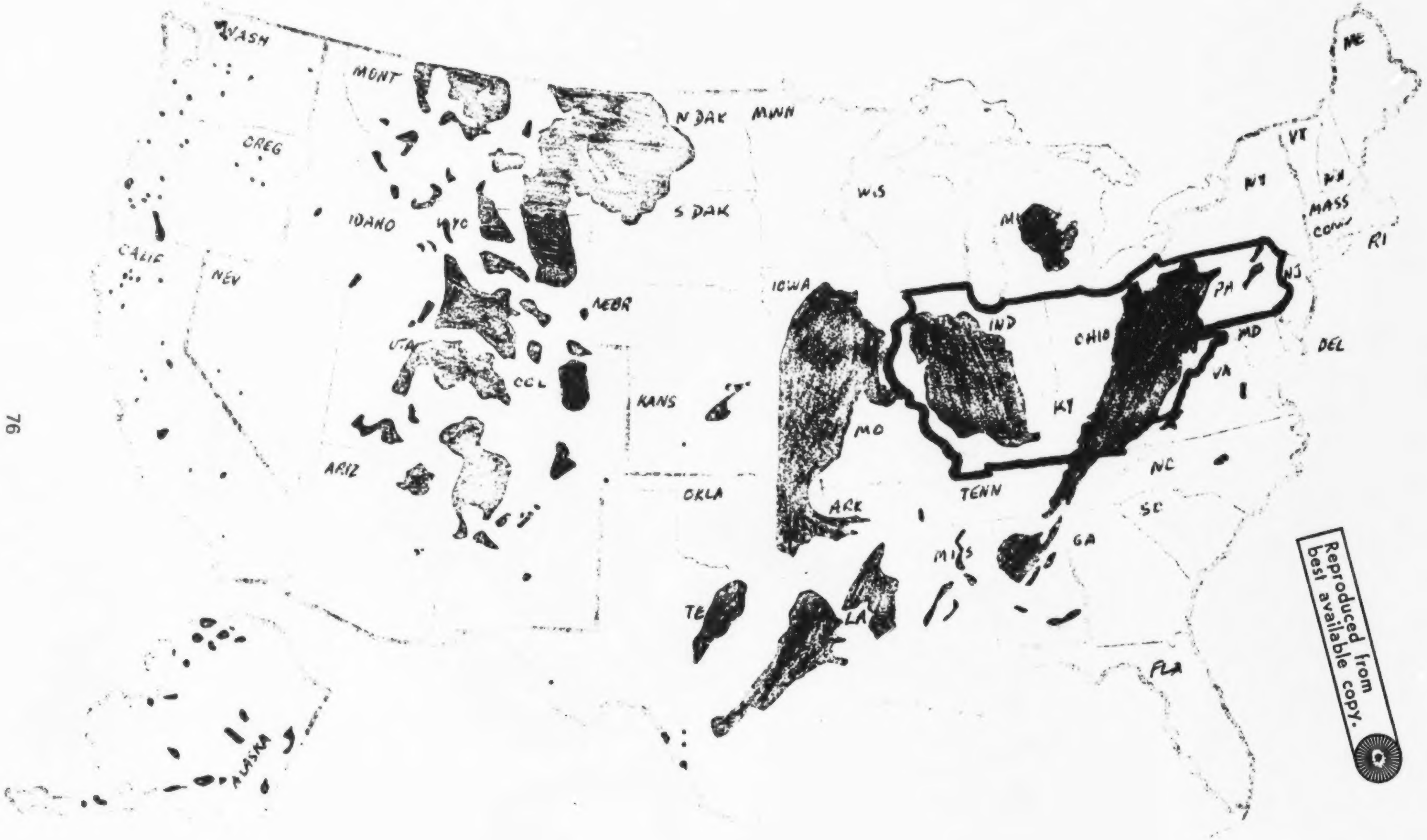
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Figure 1. Coal Fields of the United States

Table 8. Selected State Receiving Water Quality Standards

| STATE | POLLUTANT | STANDARD |
|---|---|---|
| Ohio | Toxic Substances | 0.1 TLm-48 hour |
| | Cyanide | 0.20 mg/liter |
| | Threshold Odor No.(60°C) | 24 |
| | Temperature | Seasonal limits |
| Pennsylvania | Phenol | 0.005 mg/liter |
| | Total iron | 1.5 mg/liter |
| | Total manganese | 1.0 mg/liter |
| | Threshold Odor No. (60°C) | 24 |
| | Temperature | Max. 87°F |
| West Virginia | Toxic Substances | 0.1 $Tl_m$-96 hour |
| | Cyanide | 0.025 mg/liter |
| | Phenol | 0.001 mg/liter |
| | Arsenic | 0.01 mg/liter |
| | Chromium (hexavalent) | 0.05 mg/liter |
| | Threshold Odor No. (60°C) | 8 |
| | Temperature | Seasonal limits |
| ORSANCO | Cyanide | 0.2 mg/liter |
| | Chromium (hexavalent) | 0.05 mg/liter |
| | Arsenic | 0.05 mg/liter |
| | Threshold Odor No. (60°C) | 24 |
| | Temperature | Seasonal limits |

## *Standards on Level of Pollutant Discharge*

### *Federal Performance Standards*

Regulatory limitations on pollutant discharges to the environment are of several types. Both air and water pollutions are subject to new source performance standards (NSPS) which limit emissions of particular substances from specific processes or unit operations that are newly constructed or "substantially modified" subsequent to publication of the NSPS. These standards are promulgated by the Federal Government (U.S. Environmental Protection Agency) as authorized by the 1970 Clean Air Act Amendments and the 1972 Federal Water Pollution Control Act Amendements. New source standards call for the use of "best adequately demonstrated technology," which in the case of air pollution must also take cost into account. Federal standards also apply to existing sources of water pollution. For source types specified by the EPA, effluent limitations include installation of "best pollution control technology currently available," to be installed by mid-1977, and "best available technology economically achievable," to be in place by mid-1983. There is also a congressionally mandated national goal to "zero discharge" by 1985.

For both air and water pollutants, Federal NSPS will generally impose the most stringent limitations on allowable release of pollutants to the environment. While no standards yet exist for coal conversion processes, NSPS have been promulgated or proposed for industrial processes that are related to coal conversion schemes. Petroleum refineries and coal carbonization plants offer two particularly interesting cases since they encompass many of the operations anticipated in coal conversion processing.

Table 9 indicates the air and water pollutants subject to NSPS for four industrial processes as well as new fossil-fuel-fired steam generators. Tables 10,11, and 12 give selected current standards. Proposed effluent guidelines for petroleum refineries divide the industry into six subcategories based on the raw waste load with respect to the type of

Table 9. Selected Federal New Source Performance Standards*

| Substance | Steam Generators | Petroleum Refineries | Coal Coking | Iron Making | Steel Making |
|---|---|---|---|---|---|
| AIR POLLUTANTS | | | | | |
| Particulate Matter | x | x | | x | x |
| Sulfur Oxides | x | x | | | |
| Nitrogen Oxides | x | | | | |
| Hydrocarbons | | x | | | |
| Carbon Monoxide | | x | | | |
| WATER POLLUTANTS | | | | | |
| Biochem.Oxygen Demand | | x | x | | |
| Chemical Oxygen Demand | | x | | | |
| Organic Carbon | | x | | | |
| Suspended Solids | | x | x | x | x |
| Ammonia | | x | x | x | |
| Chromium | | x | | | |
| Oils and Grease | | x | x | | |
| Phenols | | x | x | | |
| Sulfide | | x | x | x | |
| Cyanides | | | x | x | |
| Zinc | | x | | | x |
| pH | x | x | x | x | x |
| Heat | x | | | | |
| Chlorine Residual | x | | | | |
| Corrosion Inhibitors | x | | | | |
| Fluoride | | | | x | x |
| Nitrate | | | | | x |

*Refs. 20-23,30

refinery, process technology employed, and the waste severity from the operations (ref. 20). The subcategories are (a) topping, (b) low cracking, (c) high cracking, (d) petrochemical, (e) lube, and (f) integrated. Table 10 indicates the range of pollutant effluent limits over all subcategories. Typically, the lower limit applies to the most complex refinery (integrated). Note that new source refinery standards specify both the maximum effluent for any one day, and the maximum average of daily values for any period of 30 consecutive days.

The proposed NSPS limitations for byproduct coking are listed in table 11. Values for each pollutant are also specified for a 30-day average limit and a maximum daily limit (ref. 21). Comparing tables 10 and 11, one notes that the Federal effluent limitations differ in several important ways. The petroleum guidelines set different limits for different categories. More complex plant are permitted larger discharges (ranging an order of magnitude) of pollutant per unit of feedstock. All coke plants, however, are limited to a single standard, regardless of process technology or product stream. Petroleum limits are based on the quantity of feedstock processed, while coke plants are limited on basis of the product produced. While fundamentally the latter type of limit (based on product output) rewards process efficiency, the conversion efficiency of coal to coke does not vary enough to make this an effective incentive. Crude feedstock seems to have been selected as the basis for petroleum limits because refinery products are too numerous and changeable to serve as an acceptable reference basis.

enough to make this an effective incentive. Crude feedstock seems to have been selected as the basis for petroleum limits because refinery products are too numerous and changeable to serve as an acceptable reference basis.

On the air pollution side, NSPS for processes related to coal conversion apply to fewer pollutants, but are specific to individual unit operations (tables 9, 12). For fuel combustion, emissions are limited either on the basis of heat input rate (boilers) or pollutant input mass concentration (refinery plant gas). For process operations, emission limits are based principally on mass or volume concentration and opacity. One NSPS also specifies control devices.

Finally, a special class of federally mandated discharge limitation applies to substances categorized as hazardous pollutants (air lexicon), or toxic substances (water lexicon). Such substances are generally discharged in relatively few and highly localized locations, but are particularly harmful to health and require stringent control. Table 13 summarizes substances presently categorized as hazardous or toxic.

### *State and Local Regulations*

For sources or processes not covered by Federal new source performance standards (including all existing stationary air pollution sources), emission and effluent limitations are established by State and local agencies, subject to EPA approval except where such regulations are more stringent than Federal performance standards, in which case the local standard takes precedence. For example, 19 States have adopted regulations for sulfur dioxide emissions from fossil-fuel-fired steam generators that are more stringent than the new source performance standard for solid fuels (ref. 24).

An illustration of the form and variation in State air pollution regulations is provided in table 14 which shows selected particulate, $SO_2$, and $NO_x$ standards for the six eastern coal States noted earlier. For sulfur recovery plants, the Ohio regulation of 0.01 lb $SO_2$/lb S input is the most stringent mass emission regulation for plant sizes less than 900 tons/day (ref. 8), and corresponds to 99.5 percent control. This is the regulation suggested by EPA in its guidelines for State implementation plans (ref. 25). Table 15 shows emission limitations recently adopted by the State of New Mexico specifically for coal gasification plants

### Table 10

### NEW SOURCE PERFORMANCE STANDARDS FOR PETROLEUM REFINING
(pounds per 1000 barrels of feedstock)

| Pollutant | 30-Day Max** Range | One Day Max* Range |
|---|---|---|
| 5-day Biochemical Oxygen Demand (BOD5) | 1.5-6.6 | 1.85-8.2 |
| Chemical Oxygen Demand (COD) | 5.3-48.2 | 6.6-60.2 |
| Total Organic Carbon (TOC) | 1.3-9.2 | 1.6-11.4 |
| Total Suspended Nonfilterable Solids (TSS) | 0.93-4.2 | 1.2-5.2 |
| Ammonia (as N) | 0.30-2.6 | 0.40-3.4 |
| Total Chromium (CrT) | 0.023-0.106 | 0.030-0.132 |
| Hexavalent Chromium (Cr6) | 0.00046-0.0021 | 0.00058-0.0026 |
| Oils and Grease | 0.46-2.1 | 0.58-2.6 |
| Phenols | 0.0099-0.046 | 0.014-0.065 |
| Sulfide | 0.0081-0.038 | 0.013-0.059 |
| Zinc | 0.046-0.16 | 0.058-0.21 |
| pH (all categories identical) | 6.0-9.0 | 6.0-9.0 |

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[*] maximum for any one day

[**] maximum average for daily values of any period of 30 consecutive days

### Table 11

### NEW SOURCE PERFORMANCE STANDARDS FOR BY-PRODUCT COKE MAKING
(pounds per 1000 pounds of coke)

| Pollutant | One Day Max.* | 30 Day Max.** |
|---|---|---|
| Cyanides amenable to chlorination | 0.0002 | 0.0001 |
| Phenol | 0.0004 | 0.0002 |
| Ammonia (as N) | 0.0083 | 0.0042 |
| 5-day Biochemical Oxygen Demand (BOD5) | 0.0166 | 0.0083 |
| Sulfide | 0.0003 | 0.0001 |
| Oil and Grease | 0.0083 | 0.0042 |
| Total Suspended Nonfilterable Solids (TSS) | 0.0083 | 0.0042 |
| pH | 6.0-9.0 | 6.0-9.0 |

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[*] maximum for any one day

[**] maximum average of daily values for any period of 30 consecutive days

Table 12. Selected New Source Performance Standards (NSPS) for Air Pollution Sources*

| Source | Pollutant | Emissions Not to Exceed |
|---|---|---|
| STEAM GENERATORS | | |
| Fossil-fuel fired $\geq 250 \times 10^6$ Btu/hr input | Particulate Matter | 0.10 lb/$10^6$ Btu input<br>20% opacity |
| | Sulfur Dioxide | 1.2 lb/$10^6$ Btu (Solid Fuel)<br>0.8 lb/$10^6$ Btu (Liquid Fuel) |
| | Nitrogen Oxides (as $NO_2$) | 0.7 lb/$10^6$ Btu (Solid Fuel)<br>0.3 lb/$10^6$ Btu (Liquid Fuel)<br>0.2 lb/$10^6$ Btu (gaseous fuel) |
| PETROLEUM REFINERIES | | |
| Catalytic Cracking Unit Catalyst Regenerator | Particulate Matter | 0.027 gr/dscf + 0.10 lb/$10^6$ Btu aux. fuel. 30% opacity, except for 3 min/hr |
| | Carbon Monoxide | 0.050% by volume |
| Plant Gas Fuel Combustion | Sulfur Dioxide | 0.10 gr $H_2S$/dscf in fuel gas |
| Petroleum Storage Vessels | Hydrocarbons | Specified Vapor Pressure Limits and Required Control Devices |
| IRON AND STEEL INDUSTRY | | |
| Basic Oxygen Furnaces | Particulate Matter | 0.022 gr/dscf |

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*Refs. 20, 23

Table 13. Hazardous or Toxic Pollutants

| AIR* | WATER |
|---|---|
| Asbestos | Aldrin and Dieldrin |
| Beryllium | Benzidine and its salts |
| Mercury | Cadmium and its compounds |
| | Cyanide and its compounds |
| | DDE, TDE, DOD, and DOT |
| | Endrin |
| | Mercury and its compounds |
| | PCB's and mixtures of chlorinated biphenyl compounds with various percentages of chlorination |
| | Toxaphene (chlorinated camphene) |

*For specific sources only

Table 14. Selected State Emission Regulations for Six Eastern States

| | Large Combustion Units (>1000 MMBtu/hr) | | | General Process Operation (>500 ton/hr) | | Sulfur Recovery Plant |
|---|---|---|---|---|---|---|
| State | Particulate (lb/MMBtu) | $SO_2$ (lb/MMBtu) | $NO_x$ | Particulate (lb/hr) | $SO_2$ (ppm) | $SO_2$ (lb/lb S input) |
| Illinois | 0.10 | 1.0 (coal)*<br>0.8 (resid)<br>0.3 (dist.) | NSPS | 67.0 | $19.5\ P^{0.67}$ lb/hr P=tph wst rate) | N/A |
| Indiana | 0.10 | 1.2* | NSPS | Lesser of 69.0 lb/hr or 0.10 lb/1000 lb discharge at STP | | N/A |
| Kentucky | 0.10 | NSPS* | NSPS | 69.0 | 2000 | N/A |
| Ohio | 0.10 | 1.0 | NSPS | 69.0 | 500* | 0.01 |
| Pennsylvania | 0.10 | 0.3 (coal)*<br>0.2 (oil)* | N/A | Greater of 0.02 gr/scf or weight rate given for various processes | 500* | $0.32\ E^{-0.5}$ (E=long ton/day) |
| West Virginia | 0.05* | 1.6* | N/A | 21.2 - 50.0 | 2000 | 0.06 |

* Less stringent limits apply to parts of the state and/or some types or size of equipment.

Table 15. Proposed New Mexico Emission Regulations for Coal Gasification Plants

| Component | Gas-Fired Power Plant Associated with Coal Gasification Plants | Gasification Plants |
|---|---|---|
| Particulate Matter | 0.03 lb/MMBtu | 0.03 lb/MMBtu |
| Sulfur Dioxide | 0.15 lb/MMBtu | -- |
| Nitrogen Oxides | 0.20 lb/MMBtu* | -- |
| Non-methane Hydrocarbons | N/A | Nil |
| Sulfur (Vapor) | N/A | 0.04 lb/MMBtu |
| Reduced Sulfur (Sum of hydrogen sulfide, carbon disulfide, and carbonyl sulfide) | N/A | 100 ppm |
| Hydrogen Cyanide | N/A | 10 ppm |
| Hydrogen Chloride and Hydrochloric Acid | N/A | 5 ppm |
| Ammonia | N/A | 25 ppm |

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*Adopted as gas-burning equipment emission regulation

**Becomes 0.008 December 31, 1978

conversion processes, all of which require steam and power that is usually produced by an onsite boiler plant, often fired by coal or char in present designs. At the present time, the sulfur content of solid fuels proposed for boilerhouse use generally exceeds the Federal NSPS, which is equivalent to about 0.7 to 0.8 percent sulfur. The implication of present regulations, therefore, is that boiler plants utilizing solid fuels will likely require either (1) stack gas cleaning systems, (2) coal or char pretreatment, (3) fluidized bed combustion, with bed additives to control sulfur, or (4) conversion of the solid fuel to a low-Btu gas, followed by gas purification to remove sulfur as $H_2S$. Further development and demonstration of all four alternatives, however, is needed to evaluate both the technology and economics, including the economics relative to alternatives such as the use of cleaner synthetic fuel products.

Note too that existing NSPS for $SO_2$ from steam generators do not include a limitation for gaseous fuels, such as an $H_2S$-laden low-Btu gas. Such a standard, however, does exist for combustion of petroleum refinery plant gas, where $SO_2$ emissions are restricted by a limitation on $H_2S$ content of 0.10 gr/dscf (table 12). For high-Btu gasification processes, this $H_2S$ level is in fact much less stringent than chemical process requirements, which demand extremely low sulfur concentrations to avoid poisoning of methanation catalysts. However for low-Btu gas used as a boiler fuel, the refinery fuel gas standard would require more extensive desulfurization than the present solid and liquid fuel new source standards for steam generators, the most stringent of which is presently 0.8 pounds sulfur dioxide per million Btu (liquid fuels). Combustion of a low-Btu gas with a heating value of 250 Btu per cubic foot and an $H_2S$ content of 0.10 gr/dscf would be equivalent to 0.1 pounds $SO_2$ per million Btu, or nearly an order of magnitude more stringent than existing steam generator standards.

Despite this, some low-Btu gas processes for boiler fuel are being designed with gas purification systems to achieve desulfurization only to the level of new source standards for solid- and liquid-fired boilers. New regulations in this area may well be more stringent, with direct implications on the detailed design and economics of at least some coal conversion processes. Indeed, tighter regulations have already been adopted by one State (table 15).

New source particulate standards for combustion units do not present the concern occasioned by $SO_2$ standards since existing control systems such as mechanical and electrostatic collection are expected to be adequate. Nonetheless, particulate collection from boilers fired with low-sulfur char or synthetic liquid fuels has not yet been demonstrated, and such projects will have to be undertaken before the implications of new source particulate standards for steam generators can be fully assessed.

Standards for nitrogen oxides might also pose problems for auxiliary boilers. While it has been clearly demonstrated for fossil-fuel-fired boilers that $NO_x$ emissions can be substantially reduced by combustion modifications, these are more readily accomplished for gas- and oil-fired equipment than for coal-fired boilers. Also full-scale data are not yet available for boilers fired on char or synthetic liquid fuels. In the latter case, the relatively high fuel nitrogen content of some synthetic liquids would tend to worsen $NO_x$ emissions (ref. 8). Again, the precise implications of new source performance standards will have to await full-scale demonstration projects.

For particulate emissions from process unit operations not presently subject to Federal new source performance standards, table 12 provides some indication of the control levels one might expect. Most of the new source particulate standards promulgated to date, including a number of industries not shown in table 9, limit emissions to approximately 0.02 gr/dscf, or control levels typically in excess of 99 percent relative to uncontrolled processes. Applicable technologies include electrostatic precipitators, high energy scrubbers, and fabric filters, all of which have been demonstrated to achieve high degrees of particulate control.

### *Secondary Environmental Impacts*

Environmental impacts from coal conversion plants will include various secondary as well as primary impacts. An example of this is the utility (steam and power) requirement of gas-cleaning and effluent treatment systems. Here, the incremental atmospheric emissions, resulting from boilerhouse fuel consumed onsite to control process air and water pollution, further degrades the air environment.

Tradeoffs are found among pollutants discharged to a given medium as well as to different media. Many of these are well known, such as limestone scrubbing of sulfur dioxide stack gases, which can produce a potential solid waste disposal problem. Many of these tradeoffs are difficult to acknowledge in regulatory policy. This is due to both the existing institutional arrangements and the basic difficulty of weighing the relative significance of discharging different substances to different media.

Less understandably, existing regulatory policies with respect to emission standards also appear to discourage tradeoffs or incentives to minimize emissions of the *same* pollutant to the *same* medium.

Figure 2 illustrates one secondary impact. It shows the combined $SO_2$ emission rate of the sulfur recovery tailgas plus that portion of the boilerhouse stack gas resulting from steam generation for desulfurization (i.e., onsite steam-generated electric power plus direct steam requirements). Sulfur recovery efficiency is assumed to be 99.5 percent, or an $SO_2$ emission rate of 0.01 lb $SO_2$ per lb of sulfur processed. Steam is assumed to be supplied by a boiler fired with solid fuel, emitting $SO_2$ at the level of the new source performance standard (1.2 lb/million Btu). The required net energy addition is taken as 1,000 Btu per pound of steam produced, with a boiler thermal efficiency of 80 percent. This gives a heat input requirement of 1,250 Btu per pound of steam. A desulfurization system requiring 7 lb steam/lb S thus causes an incremental $SO_2$ emission at the boiler house of 0.01 lb $SO_2$/lb S, or an amount equal in magnitude to the assumed emission at the tailgas stack. The boilerhouse energy requirement in figure 2 is taken over the range of values reported for several processes desulfurizing coke oven gas (ref. 26). Low pressure coal gasification systems would likely have similar energy requirements, with less energy required for higher pressure operations.

For this example, the effective control efficiency decreases by 0.075 percent for each lb steam/lb S when additional steam requirements are accounted for. Thus, a sulfur recovery system with a nominal recovery efficiency of 99.9 percent requiring 12 lb of boilerhouse steam per pound of sulfur processed effectively operates at 0.9 percent less, or 99.0 percent.

The question of intramedia tradeoffs is also suited to this example since energy requirements for sulfur removal increase exponentially with increasing removal efficiency for many processes. Indeed, it is easy to envision cases in which a reduction in process sulfur recovery efficiency (which increases plant tailgas emissions) could reduce the *total* plant emissions (tailgas plus stack gas) because of a more substantial reduction in boilerhouse energy requirements. How significant such tradeoffs will prove to be in practice, of course, remains largely to be seen, as does the question of how such findings might influence development of future new source performance standards. Nevertheless, regulatory policies in place today would not permit a new coal conversion plant to implement a favorable tradeoff of this type, even if it were possible, if it required relaxing the NSPS for a given unit operation.

### *Implications of Multiple Regulatory Authorities*

Current institutional arrangements for implementing environmental regulatory policy call for a sharing of regulatory activities among Federal, State, and local agencies which are often charged with only a single area of environmental concern. A case study can perhaps best illustrate the potential implications of such an arrangement on coal conversion processes.

### *Catch-22: The Allegheny County Experience*

The largest byproduct coke manufacturing facility in the world is the U.S. Steel facility at Clairton, Pennsylvania, which processes about 30,000 tons of coal per day, or several times more than most coal conversion plants proposed for commercial scale. Indeed, existing coal processing plants of this size are few (of the 64 coke plants in the United States only six process more than 9,200 tpd coal).

For many years, the Clairton coke plant has operated with zero discharge of a major process wastewater by using it to quench hot coke. This complied with a Pennsylvania State regulation forbidding the discharge of certain contaminants into the waters of the State. Recently, however, the U.S. Steel entered into a consent decreee with the U.S. EPA, the State of Pennsylvania, and Allegheny County to develop a wastewater treatment system for coke plant wastes. This major change in operating practice was brought about by an enforcement action of the Allegheny County Bureau of Air Pollution Control, under a local air pollution regulation that does not permit water to be used for quenching that is not suitable for discharge to the nearest stream.

Another Allegheny County air pollution regulation prevents the water quenching of slag--a major byproduct from the production of pig iron--unless the discharge to the air of hydrogen sulfide or other contaminants is prevented. Here too, a major negotiated control program is underway to solve a problem which again bears similarities to aspects of at least some coal conversion processes.

The implication of cases such as this is to strongly underscore the need for an early and thorough assessment of process environmental impacts in order to minimize or avoid future problems. Similarly, the early and coordinated involvement among local, State

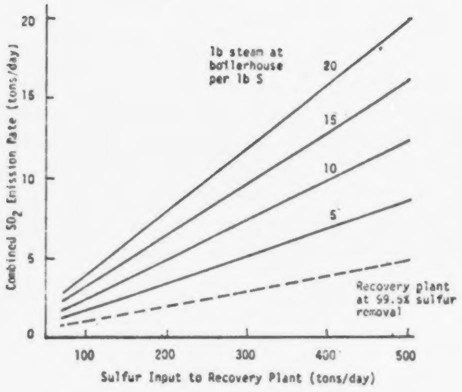

Fig. 2 Total $SO_2$ Emissions from Sulfur Recovery Tailgas Plus Boilerhouse Steam for Sulfur Recovery (Boiler in compliance with NSPS for solid fossil fuel)

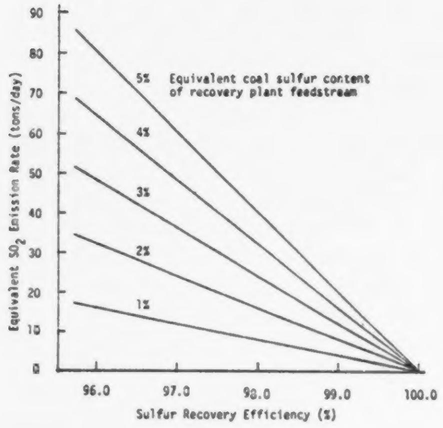

Fig.3 Influence of Coal Type and Control System Efficiency on Equivalent $SO_2$ Emissions from a Sulfur Recovery Plant at a 20,000 Ton/Day Coal Conversion Facility

and Federal regulatory authorities is essential for the efficient implementation of environmental regulatory policies.

## IMPLICATIONS OF ENVIRONMENTAL QUALITY STANDARDS

### *Ambient Air Quality Standards*

As noted earlier, national ambient air quality standards (AAQS) for six major air pollutants have been promulgated by the U.S. Environmental Protection Agency (table 5). Individual States have promulgated other AAQS that apply locally, and may be more encompassing or more stringent than the national standards.

The implications of ambient air quality standards on coal conversion processes go directly to questions of plant siting. One must consider all regional sources of air pollution, and the way in which they interact with the coal conversion plant to affect ambient air quality at any location. This involves concern for the geographical distribution of sources, pollutant mass emission rates, discharge stack heights, process parameters and local or regional meteorology. Even where there is ready compliance with all applicable emission standards, the added constraint of air quality standards can profoundly restrict the design flexibility and siting of a particular facility.

### *Coupling of Emissions to Air Quality*

A simple illustration draws on the case of present (State) regulations for sulfur recovery plants which specify a sulfur recovery efficiency by defining the maximum allowable mass emission rate of $SO_2$ per mass of sulfur input. Figure 3 indicates how the total mass emission rate of sulfur dioxide varies with sulfur recovery efficiency for five different coals at a plant with a total coal input rate of 20,000 tons per day. Plants processing different coals in compliance with a specified sulfur recovery efficiency emit different total masses of sulfur dioxide, which can have substantially different impacts on environmental quality. At a control efficiency of 99.5 percent, a plant processing a Pittsburgh seam coal with a sulfur content near 4 percent would emit approximately 8 tons/day of $SO_2$, while the same plant operating on an eastern Kentucky coal of 1 percent sulfur would emit about 2 tons/day. Although both plants would be in compliance with the applicable $SO_2$ emission standard, the former case could result in up to a four-fold degradation of air quality relative to the latter case.

A quantitative estimate of the ambient concentrations resulting from a specified pollutant mass emission rate can be obtained with diffusion

Table 16. Adjusted New Source Performane Standards for By-Product Coke Making and Petroleum Refining (30-day Maximum)

(pounds of pollutant per $10^{12}$ Btu feedstock)*

| Pollutant | Petroleum Refineries | By-Product Coke Making |
|---|---|---|
| BOD5 | 230-1015 | 477 |
| COD | 815-7415 | N/A |
| TOC | 200-1415 | N/A |
| TSS | 143-646 | 242 |
| Ammonia (as N) | 46-400 | 242 |
| Total Chromium | 3.5-16 | N/A |
| Hexavalent Chromium | 0.07-0.32 | N/A |
| Oil and Grease | 71-323 | 242 |
| Phenols | 1.5-7.1 | 12 |
| Sulfide | 1.2-5.8 | 5.8 |
| Zinc | 7.1-25 | N/A |
| Cyanide amenable to Chlorination | N/A | 5.8 |

*Assumes heating values of 6.5 MMBtu/bbl for crude oil and 12,000 Btu/lb for coal, with a coke yield of 0.69 lb coke/lb coal.

N/A not applicable

and auxiliary boilers, in anticipation of two Lurgi-based gasification plants scheduled for construction. These are presently the only State regulations directed at coal conversion processing.

For water pollutants, State and local standards will generally not be as stringent as Federal effluent guidelines, although some exceptions can be expected. Pennsylvania, for example, presently permits no measurable discharge of cyanide or phenol into waters of the Commonwealth.

## IMPLICATIONS OF PRESENT EMISSION AND EFFLUENT STANDARDS FOR FUTURE COAL CONVERSION PROCESSES

Current regulatory activities suggest that coal conversion processes will be subject to separate emission and effluent limitations in much the same way that new sources in various industrial categories are presently regulated. Although this may not be the optimal regulatory framework in which to minimize the environmental impact of coal conversion factilities, current policy points strongly in this direction, at least for the immediate future. It is useful, then, to examine the implcations that existing emission and effluent limitations will or could have on coal conversion processes.

### *Implications of Existing National Effluent Standards*

Some insights as to the nature of effluent standards for coal conversion processes can be obtained by comparing the new source performance standards for the related industries of byproduct coke making and petroleum refineries, shown earlier (tables 10 and 11). The two sets of standards may be compared directly by expressing them on the common basis of energy input of the process feedstock (table 16). For this comparison, crude oil is assumed to have a heating value of 6.5 million Btu/bbl. Standards for coke making based on coke produced are transformed to a heat input basis assuming each ton of coal yields 0.69 tons of coke (the national average), and assuming coal has a heating value of 12,000 Btu/lb.

The range of the limits for petroleum refineries in table 16 again reflects the philosophy of setting separate limits that depend on process technology and product mix for refineries. No such range exists for coke making. However, with the exception of the phenol limit, which is higher for coking, and cyanide which is not explicitly limited for refineries, all coke industry limits fall within the range of the petroleum refinery limits. While these relationships may be fortuitous, they may also suggest a basis for estimating potential effluent standards for coal conversion processes. Nonetheless, since the suggested limitations in fact range over an order of magnitude for some pollutants, the capability and cost of achieving specific effluent guidelines will still depend strongly on the actual performance of individual processes.

In this regard, it is also unclear whether one might expect a range of regulations for coal conversion effluents, perhaps based on output products, in such a way as to reward process efficiency. It is similarly uncertain whether more complex plants will be subject to "end-of-the-pipe" effluent standards, which permit internal decisions for controlling effluent compositions, or to separate standards for selected processes or unit operations. Byproduct coke making, for example, is often an integral part of a steel manufacturing plant, yet its wastewaters are regulated separately from those of iron or steel making processes which discharge the same pollutants into the same pipe at the same plant.

### *Implications of Existing National Emission Standards*

Of the several existing new source standards for air pollutants, those for fossil-fuel-fired steam generators (table 12) will have a direct bearing on coal modeling techniques. Figure 4 shows isopleths of annual average groundlevel $SO_2$ concentration for a hypothetical coal conversion plant located in southwestern Pennsylvania, predicted by the Air Quality Display Model (ref. 27). The plant is assumed to have two sources of $SO_2$ emissions. One is the stack gas of a boiler operating in compliance with the NSPS of 1.2 lb $SO_2$/million Btu input for solid fossil fuel at a heat input rate of 2,500 million Btu/hour (yielding an emission of 36.0 tons of $SO_2$/day). The second is the tailgas stream of a sulfur recovery plant processing 500 short tons per day of sulfur at a required recovery efficiency of 99.9 percent (yielding an $SO_2$ emission of 1.0 ton/day). The two sources are assumed to be at approximately the same location, with effective stack heights of 400 feet and 200 feet, respectively. For these conditions, the predicted maximum annual average $SO_2$ concentration is 37 $\mu g/m^3$, or nearly half the national primary standard, without considering the added contribution of nearby or background sources. Increases in the assumed stack heights, however, would significantly decrease groundlevel concentration.

Figure 5 isolates the annual $SO_2$ concentration due only to tailgas emissions plus that part of boilerhouse emissions required for sulfur recovery, where steam requirements are assumed to be 12 lb/lb S

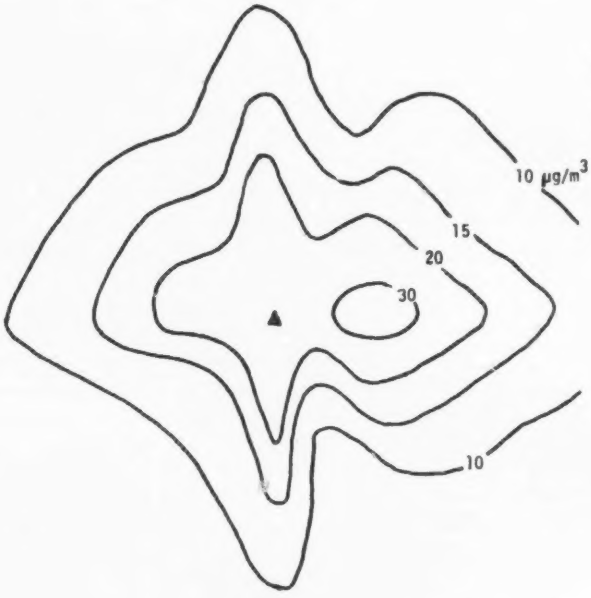

Fig.4. Annual Average $SO_2$ Concentration ($\mu g/m^3$) for a Hypothetical Coal Conversion Plant (▲ indicates source location)

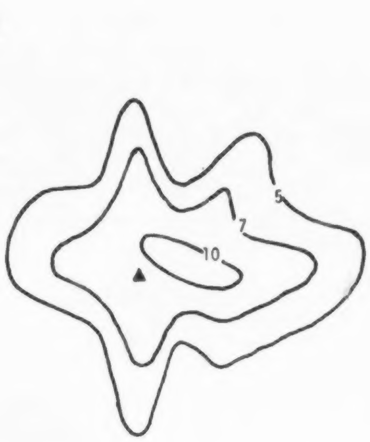

Fig.5. Annual Average $SO_2$ Concentration ($\mu g/m^3$) Due Only to Combined Effect of Tailgas Emission Plus Boilerhouse Steam for Sulfur Recovery System

(contributing 9.0 tons/day to total $SO_2$ emission). Approximately 80 percent of the peak concentration here is due to the boilerhouse $SO_2$ emission.

### *Implications of Nondegradation Standards*

The present Federal mandate to prevent "significant deterioration" of clean air is a particularly crucial issue in terms of the implications of environmental policy on coal conversion processes. Indeed, the tentative levels of incremental degradation shown earlier in table 7 would preclude operation of the hypothetical plant of figure 4 in many areas of the country. Alternately, they would demand more stringent emission controls, alternative cleaner fuels and/or taller stacks to reduce ambient concentrations adequately. Nondegradation policies of the type presently being contemplated, therefore, may well pose the most critical constraints on the siting and design of coal conversion facilities.

### *Impacts on Receiving Water Quality*

It was indicated earlier that receiving water quality standards are in practice not as strongly coupled to discharge standards as are air quality and emission standards. Nonetheless, water quality considerations are indeed present and must be considered.

One assessment of the environmental impact of water pollutant discharge from coal conversion facilities may be obtained by examining the relative quantities of emissions of the same pollutant from all sources in a given region. For water problems, it is most reasonable to examine a watershed. Table 17 lists the distribution of refineries and byproduct coke plants in a six-State region that affects the watershed primarily of the Ohio River. The list includes all facilities in the State, not just those discharging into the Ohio River and its tributaries. Nevertheless, the methodology provides a key to assessment. This six-State region holds more than one-fifth of the U.S. petroleum refinery crude capacity, as well as slightly less than two-thirds of the country's byproduct coke capacity.

Table 18 estimates the total regional burden of selected pollutants for a situation in which six new 20,000 ton/day coal conversion plants are assumed to be sited in the region (an average of one per State), with all sources complying with the NSPS indicated in the table. In terms of total effluent quantities, six plants would contribute as much as either coking or refineries. This comparison does not, of course, deal with the more important problem of the local impact of each individual facility, which is expected to be considerable when one considers that such plants are of size comparable to the Clairton coke plant. However, the estimate of total regional emissions does provide some perspective of the impact of new coal conversion facilities for this eastern area.

## SUMMARY AND CONCLUSIONS

These discussions have examined detailed aspects of current environmental regulatory policy for air and water pollution control as they are likely to bear on coal conversion processes presently under development in this country. The discussions focused on the types and levels of controls likely to be required, and attempted to illustrate existing or potential areas of conflicts among standards for air and water discharges, and levels of environmental quality.

The analysis suggests a series of questions requiring further consideration and study in the development of coal conversion plant regulatory policies:

- To what extent is a multimedia (air, water, land, etc.) approach to environmental control necessary? How do we evaluate cross-media tradeoffs?
- Within a single medium, is it necessary to allocate waste loads according to specific processes or unit operations? Can a "total plant" standard lead to less severe environmental degradation?
- How should plant size and product mix enter into the regulatory picture? Can incentives be structured to reward process efficiencies that reduce environmental impact?
- Can the existing U.S. regulatory structure be modified to streamline policy formulation and avoid or minimize jurisdictional conflicts (Federal vs. State vs. local; air vs. water vs. other)?

Clearly, these questions are by no means unique to coal conversion processes, but in fact could be applied to virtually any of our existing industries. What makes this particularly relevant to coal conversion, however, is that we are in this case speaking of a potentially large-scale industry whose environmental impacts can be significant, but which is at present less constrained and encumbered relative to other industries, hence, better able to respond to environmental concerns.

In large part, the key to success will lie in the early recognition of environmental factors, and in the integration of environmental and process considerations. Indeed, the substance of the case for

Table 17. Distribution of Coke Plants and Petroleum Refineries for the Six Major Eastern Coal States

| State | No. of Refineries | 1971 Refinery Capacity* (1000 bbl/day) | No. of Coke Plants | No. of Ovens |
|---|---|---|---|---|
| Illinois | 10 | 908 | 5 | 455 |
| Indiana | 9 | 558 | 6 | 2361 |
| Kentucky | 3 | 163 | 1 | 146 |
| Ohio | 8 | 572 | 13 | 1796 |
| Pennsylvania | 12 | 683 | 12 | 3491 |
| West Virginia | 2 | 11 | 3 | 668 |
| | 44 | 2895 | 40 | 8917 |
| Total USA | 247 | 13000 | 64** | 4255 |

*Ref. 29.
**The 64 coke plants process about 82 million tons of coal per year.

Table 18. Estimated Total Wastewater Emissions of Selected Pollutants for a Six-State Region

(pounds per day based on 30-day average NSPS limits)

| Pollutant | Refineries* | Coke Plants** | Coal Conversion Plants*** |
|---|---|---|---|
| Phenols | 30-138 | 39 | 33 |
| Ammonia | 900-7,800 | 1638 | 1378 |
| BOD5 | 4,500-145,000 | 2910 | 2756 |
| TSS | 2,790-12,600 | 1638 | 1378 |
| Oil & Grease | 1,380-6,300 | 1638 | 1378 |

*Assumes 22 percent (3 million bbl/day) of U.S. crude flow is processed in the six-state region with range of emissions depending on process technology mix.
**Assumes 63 percent (52 million tons/yr of coal) of U.S. by-product coke is processed in the six-state region.
***Assumes six 20,000 ton/day plants (120,000 tons of coal per day, total) operating at NSPS limits for coke plants over the six-state region.

careful assesssment and management of the environmental problems of coal conversion processes was summarized recently in a report on the "Nation's Energy Future" (ref. 1) and bears repeating here:

> "Disruption of the energy program can be prevented by anticipating potential problems related to each technology and by determining as rapidly as possible the effects on health, ecosystems, and society. Perhaps the largest barrier to be faced is the need to convince energy-related technologists and planners that this seemingly distractive commitment must be made at the outset to prevent very major disruptions in energy production."

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# ENVIRONMENTAL ASPECTS OF EL PASO'S BURNHAM I COAL GASIFICATION COMPLEX

**Cecil R. Gibson,**
**Gene A. Hammons, and**
**Don S. Cameron***

*Abstract*

*The El Paso Natural Gas Company plans to build a fuel conversion complex, utilizing proprietary technology supplied by Lurgi Mineraloltechnik GMBH and others. This complex will convert coal to high-Btu pipeline quality gas. The basic processes utilized in the complex will be reviewed briefly.*

*The quality, quantity, and composition of the effluent streams from the complex will be presented. Over 98 percent of the sulfur in the feed to the complex will be recovered. Maximum use of water will be achieved by recycling all reclaimable internal effluents. Aqueous waste streams which cannot be recycled will be discharged to evaporation ponds.*

*El Paso's Burnham I Coal Gasification Complex has been designed to meet all applicable environmental standards. Extensive studies designed to obtain data on the existing environment are being conducted to add to the existing knowledge of the area.*

## INTRODUCTION

Coal is the most abundant source of fossil fuel energy in the United States. Combustion of coal often results in unacceptable levels of pollutants; therefore, various methods of utilizing coal while producing minimum pollutants are being examined.

The conversion of coal into a gas which is interchangeable with natural gas (synthetic natural gas or SNG) is one means of producing clean energy from coal and producing minimum air and water pollutants. El Paso Natural Gas Company plans to build a plant in Northwestern New Mexico which will convert 28,254 tons of coal per day into 288 million scfd of SNG. The plant will be based on Lurgi technology. This facility, named the Burnham Coal Gasification Complex, is estimated to cost about \$605 million, based on mid-1973 dollars. The plant, including all utilities, will have a gross investment of \$491 million, and the mine and coal preparation facilities will have a gross investment of \$114 million. The product gas from this plant, which will have a heating value of 954 Btu/cf, will be delivered to El Paso's mainline system at a 25-year average cost of about \$1.17/mcf.

Oxygen-blown Lurgi gasifiers, as illustrated in figure 1, will be used to produce a medium-Btu (appoximately 320 Btu/scf) gas from coal. Since coal is deficient in hydrogen (as compared to methane), large quantities of steam are also utilized in the gasifier as a hydrogen source. The primary processing units required to upgrade the medium-Btu gas to SNG are illustrated in figure 2.

Airblown Lurgi gasifiers will be used to produce a fuel gas (approximately 190 Btu/scf). This gas will be used to fuel the gas turbines and steam boilers and thus produce the electricity and steam requirements for the plant.

## CONTROL OF AIR POLLUTANTS

The State of New Mexico air pollution emission regulations with which the Burnham Complex must comply are illustrated in table 1.

### *Sulfur Control*

A sulfur balance around the High-Btu Gas Production sections of the Burnham Complex is shown in figure 3. The proposed New Mexico regulations, both on total sulfur and on reduced sulfur, are achieved.

The bulk of the inlet sulfur with the coal, approximately 95 percent is converted to $H_2S$ in the gasifier. Approximately equilibrium amounts of $H_2S$, COS, $CS_2$, mercaptans, and the thiophenes ($C_4H_4S$) are formed. The percentage of the sulfur in the input coal which is converted to each of the gaseous organic sulfur compounds is given in table 2. The other organic sulfur is thus primarily mercaptans, since the equilibrium constants for the formation of $CS_2$ and thiophenes are fairly small.

Before any of the sulfur compounds are removed, a portion of the raw synthesis gas is passed through a Lurgi Crude Gas Shift Unit. The gas quantity through

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FIGURE 1

THE LURGI GASIFIER

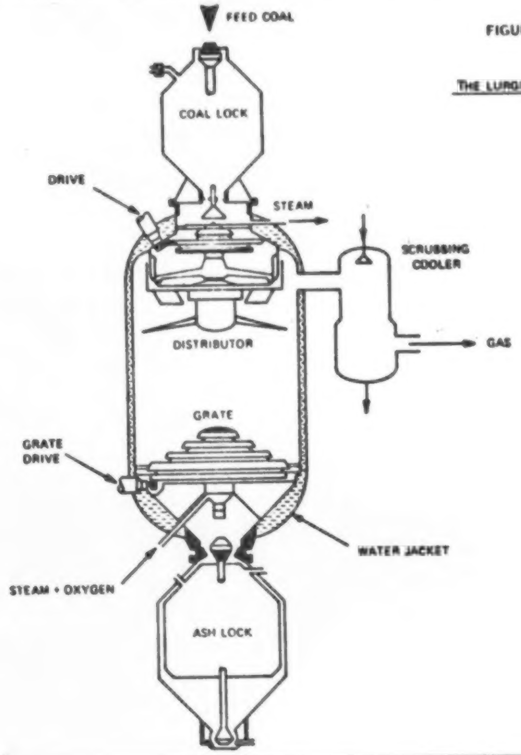

FEED COAL
COAL LOCK
DRIVE
STEAM
SCRUBBING COOLER
DISTRIBUTOR
GAS
GRATE
GRATE DRIVE
WATER JACKET
STEAM + OXYGEN
ASH LOCK

FIGURE 2

PROCESSES REQUIRED TO UPGRADE RAW GAS (320 BTU/SCF) TO SNG (984 BTU/SCF)

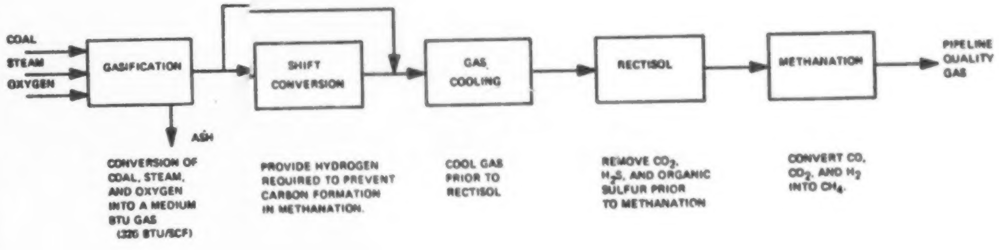
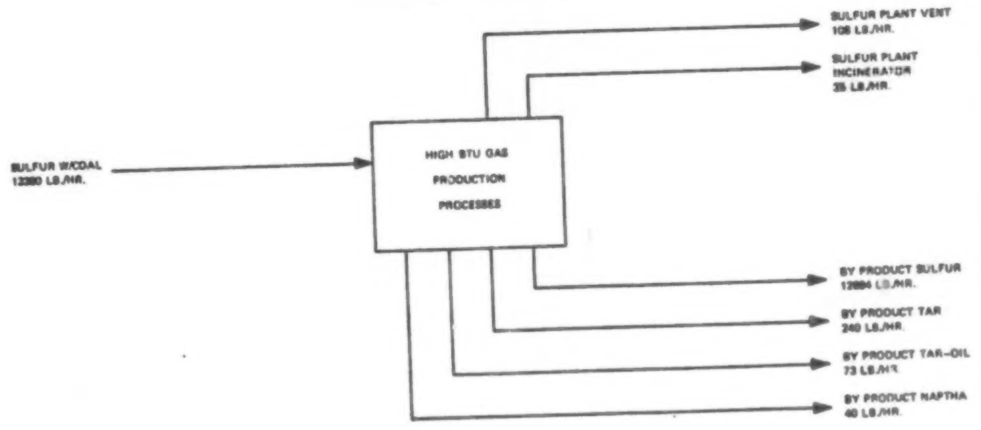

COAL
STEAM
OXYGEN
GASIFICATION
SHIFT CONVERSION
GAS COOLING
RECTISOL
METHANATION
PIPELINE QUALITY GAS
ASH
CONVERSION OF COAL, STEAM, AND OXYGEN INTO A MEDIUM BTU GAS (320 BTU/SCF)
PROVIDE HYDROGEN REQUIRED TO PREVENT CARBON FORMATION IN METHANATION.
COOL GAS PRIOR TO RECTISOL
REMOVE $CO_2$, $H_2S$, AND ORGANIC SULFUR PRIOR TO METHANATION
CONVERT CO, $CO_2$, AND $H_2$ INTO $CH_4$.

Table 1. State of New Mexico air pollution emission regulations

| | |
|---|---|
| Gas-fired boilers | |
| Sulfur dioxide emissions | 0.16 lb/MM Btu fired (LHV basis) |
| Nitrogen dioxide emissions | 0.20 lb/MM Btu fired (lhv Basis) |
| Particulate emissions | 0.03 lb/MM Btu bired (LHV basis) |
| Coal gasification plant | |
| Sulfur emissions | 0.03 lb/MM Btu fired (LVH basis) |
| Hydrogen sulfide, carbon disulfide, and carbonyl sulfide emissions | 100 ppm by volume (total) |

FIGURE 3

SULFUR BALANCE FOR MANUFACTURE OF HIGH BTU GAS


SULFUR PLANT VENT
108 LB./HR.
SULFUR PLANT INCINERATOR
35 LB./HR.
SULFUR W/COAL
12380 LB./HR.
HIGH BTU GAS PRODUCTION PROCESSES
BY PRODUCT SULFUR
12884 LB./HR.
BY PRODUCT TAR
240 LB./HR.
BY PRODUCT TAR-OIL
73 LB./HR.
BY PRODUCT NAPTHA
40 LB./HR.

NOTE: 1) ALL FIGURES ARE IN LB./HR. OF SULFUR

2) EMISSION CALCULATES AS 0.00792 LBS/MM BTU OF INLET COAL

3) SULFUR PLANT VENT IS REDUCED SULFUR WITH CONCENTRATION LESS THAN 100 VPPM

Table 2. Estimated quantities of sulfur compounds in the product gas from a Lurgi gasifier

| Compound | % of sulfur in input coal which appears in product gas as |
|---|---|
| $H_2S$ | 95.0 |
| COS | 2.4 |
| $CS_2$ | 0.3 |
| Mercaptans | 2.0 |
| Thiophenes | 0.3 |
| Total | 100.0 |

shift is set by the $H_2$ to CO ratio required in methanation, and, for the Burnham Complex, approximately 50 to 60 percent of the total gas is shifted. In the Shift Unit, the unreacted steam is used to produce $H_2$ by water-gas shift reaction:

$$CO + H_2O \rightleftharpoons H_2 + CO_2 \tag{1}$$

Since organic sulfur may be hydrolyzed to $H_2S$ and $CO_2$ at elevated temperatures (300° to 750°F), that portion of the gas which goes through shift has essentially equilibrium conversion of the organic sulfur. Table 3 illustrates the approximate equilibrium conversions of various organic sulfur compounds to $H_2S$ at approximately 720°F.

The converted gas from shift and from the crude gas bypass are recombined and cooled prior to the gas purification step. During this cooling step, gas liquor is condensed which contains water and a number of other coal carbonization products. Following cooling, the raw synthesis gas is purified in a Lurgi Rectisol unit. In this unit, the $CO_2$, $H_2S$, and organic sulfur compounds are removed from the gas by absorption in cold methanol. The Rectisol process has the following advantages for this application:

(1) It removes saturated and unsaturated hydrocarbons without contaminating the solvent beyond regeneration.

(2) It removes sulfur compounds, both $H_2S$ and organic sulfur compounds, to less than 0.2 vppm; a Rectisol unit at South African Coal, Oil, and Gas Corporation (SASOL) has achieved sulfur removal to 0.007 vppm.

(3) It effectively removes HCN.

Table 3. Equilibrium organic sulfur conversion in shift unit

| Compound | % conversion to $H_2S$ |
|---|---|
| COS | 63 |
| $CS_2$ | 100 |
| RSH | 100 |
| $C_2H_2S$ | 100 |

(4) The methanol solvent will not contaminate the methanation catalyst.

(5) It is currently in use to treat the gas from Lurgi pressure gasification. At the SASOL plant, sulfur-sensitive Fisher-Tropsch catalyst are utilized immediately following the Rectisol unit.

In the Burnham Complex, the offgas from the Rectisol solvent regeneration section has the approximate composition shown in table 4. The concentration of $H_2S$, as shown in table 4, is far too low to be used as a Claus plant feed. Normally, the feed to a Claus unit contains a minimum of 12 percent $H_2S$. Although this stream could be concentrated, economic studies have indicated that it would be better to use the Stretford Process in the

Table 4. Rectisol offgas composition

| Component | Mol % |
|---|---|
| $CO_2$ | 97.63 |
| $H_2S$ | 0.75 |
| $C_2H_4$ | 0.24 |
| CO | 0.07 |
| $H_2$ | 0.43 |
| $CH_4$ | 0.56 |
| $C_2H_6$ | 0.32 |
| $N_2$ + A | -- |
| Total | 100.00 |

Burnham Complex. This process converts the $H_2S$ and a portion of the organic sulfur to elemental sulfur by the following reactions:

$$H_2S + Na_2Co_3 \rightleftarrows NaHS + NaHCO_3 \qquad (2)$$

$$HS^- + V^{(S+)} \rightleftarrows S + V^{(4+)} \qquad (3)$$

$$V^{(4+)} \rightleftarrows V^{(S+)} \text{ Reduced ADA} \qquad (4)$$

$$\text{Reduced ADA} + O_2 \rightleftarrows \text{ADA} + H_2O \qquad (5)$$

This process will give the Burnham Complex approximately 99 percent overall sulfur recovery so as to meet all applicable sulfur emission regulations.

Coal-fired boilers will not be used in the Burnham Complex. Thus, the problem of finding a workable stack gas scrubbing system will be avoided. A power and steam generating system using airblown Lurgi gasifiers to fuel both gas turbines and waste heat boilers will be used. Since the sulfur in the coal is converted primarily to $H_2S$ in a Lurgi gasifier, high sulfur removal efficiency can be attained.

A sulfur balance around the steam and power generation system for the Burnham Complex is illustrated in figure 4. The expected emissions are 0.13 lb/$10^6$ Btu, considerably below the New Mexico and Federal regulations of 0.16 lb/$10^6$ Btu.

The Stretford process will be used to remove the $H_2S$ from the fuel gas. This Stretford absorber will operate at about 250 psig, as opposed to atmospheric operation of the Stretford absorber in the high-Btu gas production area. The chemical reactions occurring are the same as previously described.

### *Particulate Control*

The coal charged to a Lurgi gasifier is typically in the size range of 1½ X ¼ inches. Any of the fine coal which is blown out of the gasifier by the upflowing gas is removed from the gas in a water scrubber immediately following the gasifier. This coal dust tends to collect in the condensed tar, which is recycled to the gasifier. Particulate emissions from the processing area are thus negligible.

Coal gasification is a particularly attractive means of controlling particulate emissions in the utilization of low-sulfur Western coal. Precipitator efficiency has been demonstrated many times to be very poor when operating on low-sulfur coals.

Particulate emissions in the coal crushing and conveying areas will be kept to a minimum. The coal on the conveyors will be sprayed with water at transfer points to minimize particulate emissions.

### *Nitrogen Oxide Control*

Fuel nitrogen has been reported (refs. 1,2) to be more reactive for the production of nitrogen oxides ($NO_x$ control techniques, such as two-stage firing, have been reported (refs. 1,2) to be less effective on nitrogen-containing fuels, such as coal.

FIGURE 4

SULFUR BALANCE FOR MANUFACTURE OF LOW BTU FUEL GAS

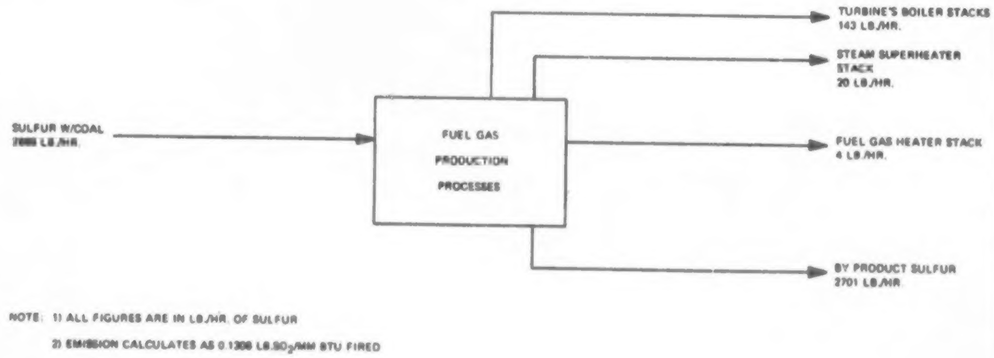
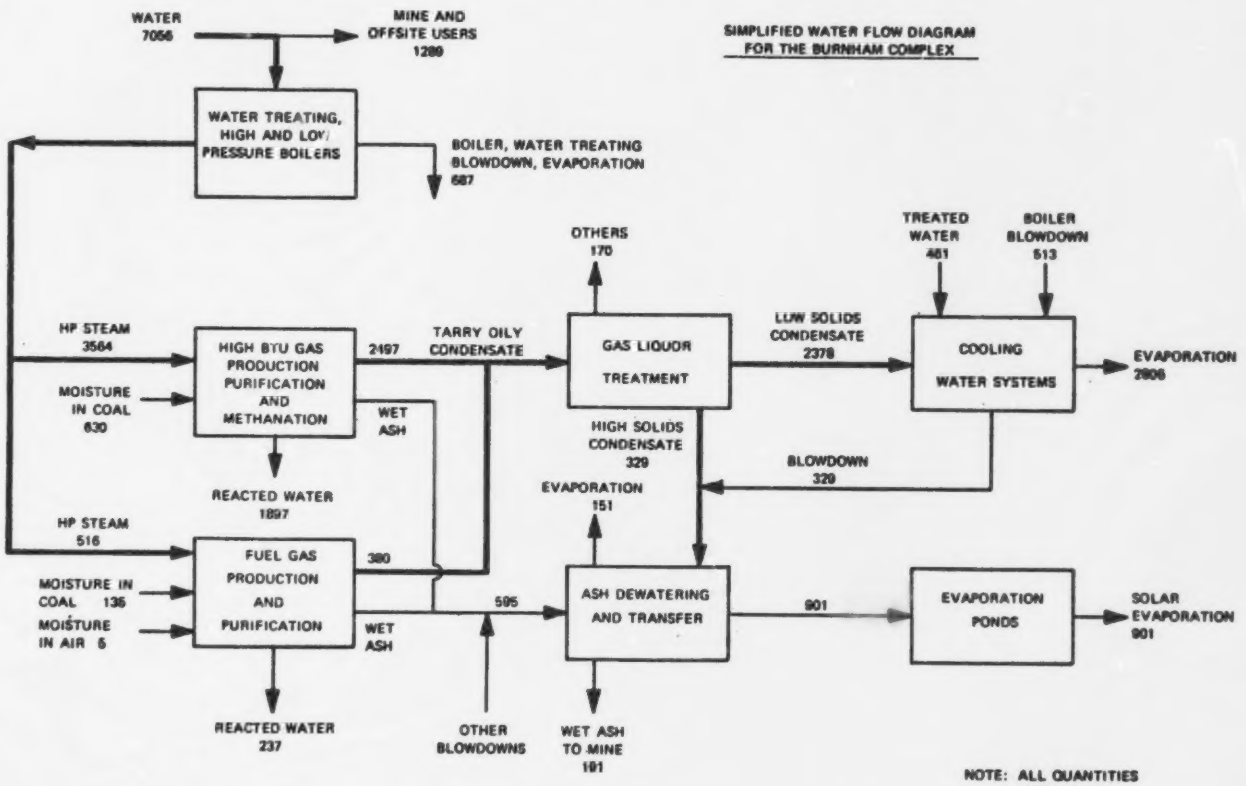


FIGURE 5

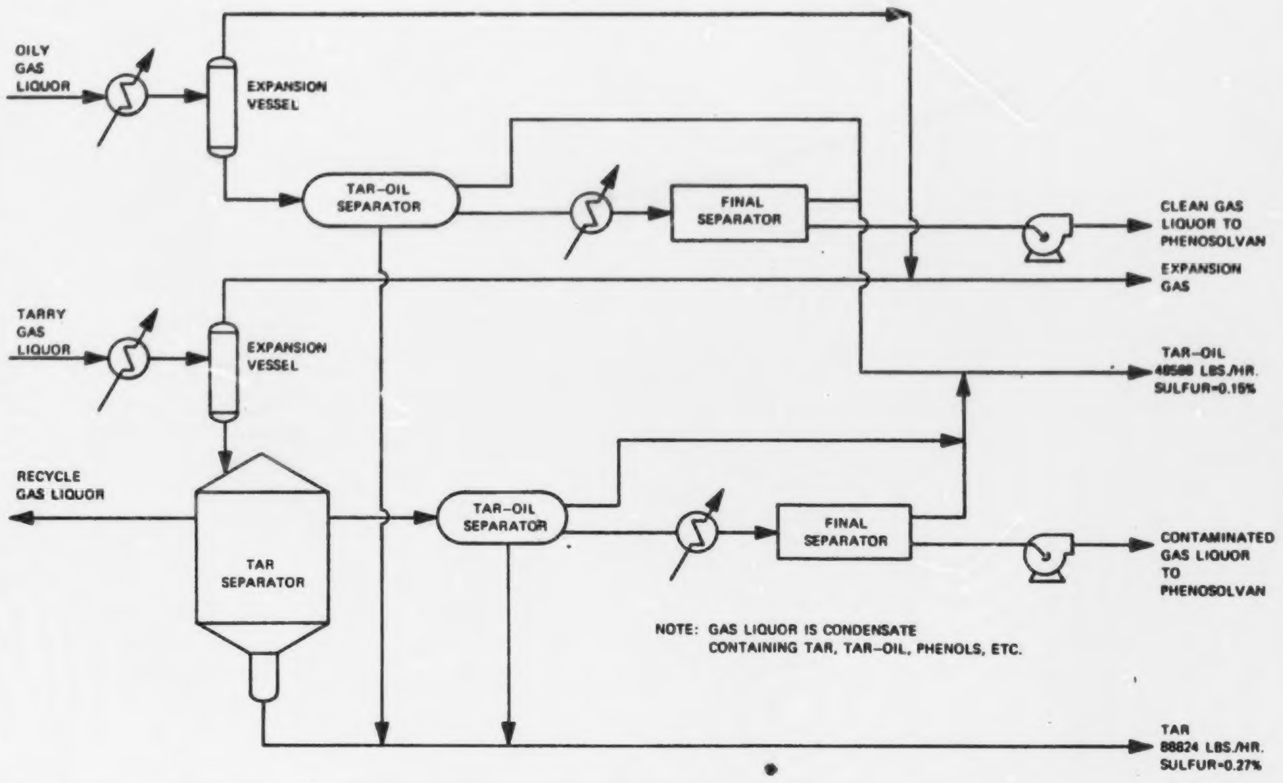
SIMPLIFIED WATER FLOW DIAGRAM FOR THE BURNHAM COMPLEX


WATER 7056
MINE AND OFFSITE USERS 1289
WATER TREATING, HIGH AND LOW PRESSURE BOILERS
BOILER, WATER TREATING BLOWDOWN, EVAPORATION 687
HP STEAM 3564
MOISTURE IN COAL 630
HIGH BTU GAS PRODUCTION PURIFICATION AND METHANATION
REACTED WATER 1897
2497
TARRY OILY CONDENSATE
WET ASH
HP STEAM 516
MOISTURE IN COAL 136
MOISTURE IN AIR 5
FUEL GAS PRODUCTION AND PURIFICATION
REACTED WATER 237
380
WET ASH
595
OTHER BLOWDOWNS
OTHERS 170
GAS LIQUOR TREATMENT
HIGH SOLIDS CONDENSATE 329
EVAPORATION 151
ASH DEWATERING AND TRANSFER
WET ASH TO MINE 191
LOW SOLIDS CONDENSATE 2378
TREATED WATER 481
BOILER BLOWDOWN 513
COOLING WATER SYSTEMS
EVAPORATION 2806
BLOWDOWN 329
901
EVAPORATION PONDS
SOLAR EVAPORATION 901

NOTE: ALL QUANTITIES ARE IN GPM

FIGURE 6

TAR AND TAR-OIL SEPARATION SYSTEM


OILY GAS LIQUOR
EXPANSION VESSEL
TAR-OIL SEPARATOR
FINAL SEPARATOR
CLEAN GAS LIQUOR TO PHENOSOLVAN
EXPANSION GAS
TARRY GAS LIQUOR
EXPANSION VESSEL
TAR-OIL 40588 LBS./HR. SULFUR-0.15%
RECYCLE GAS LIQUOR
TAR SEPARATOR
TAR-OIL SEPARATOR
FINAL SEPARATOR
CONTAMINATED GAS LIQUOR TO PHENOSOLVAN
NOTE: GAS LIQUOR IS CONDENSATE CONTAINING TAR, TAR-OIL, PHENOLS, ETC.
TAR 88824 LBS./HR. SULFUR-0.27%

The nitrogen in coal is converted primarily to $NH_3$ in a Lurgi gasifier. In the fuel gas production system for the Burnham Complex, most of the $NH_3$ will be removed with the condensate prior to the Stretford unit. The inlet temperature to the Stretford unit is 90°F. Thus, essentially no fuel nitrogen or ammonia will be combusted in the gas turbines or waste heat boilers. The absence of fuel nitrogen combined with the reduced flame temperatures (both in the turbine and waste heat boiler), due to the low heating value of the fuel gas, will allow us to achieve the New Mexico and Federal regulation of 0.20 lb/$10^6$ Btu HHV (as $NO_2$).

## CONTROL OF WATER POLLUTANTS

Steam in excess of that required for reaction is generally supplied to a Lurgi gasifier. This excess steam is required to maintain the desired temperature profile in the gasifier. The unreacted steam exits the gasifier with the raw gas. A part of the excess steam is required as a reactant in the shift conversion unit, while the remainder is condensed from the gas, along with the tar, tar oil, ammonia, phenols, other organic compounds, and acid gases.

In the Burnham Complex, the condensed excess steam will be purified to the extent required for use as cooling tower makeup water. A simplified water balance for the Burnham Complex is shown in figure 5. Approximately 2,379 gpm will be purified and reused as cooling tower makeup. About 329 gpm of condensate will be separated because of its high halogen, phenol, and other organic compound concentrations and sent to solar evaporation ponds. This high solids condensate is estimated to contain a high percentage of the chloride and fluoride in the coal. This percentage is a strong function of the temperature level at which the separation is made. By separating the halogens, the quality of the remainder of the "clean" condensate (2,379 gpm) is considerably improved for usage as cooling tower makeup water.

Boiler blowdown and cooling tower blowdown are also sent to solar evaporation ponds after being used as sluicewater in the ash-handling system.

Water reclamation techniques are being examined. Any of the water presently being sent to evaporation ponds will be reclaimed if economically feasible.

### *Tar and Tar Oil Removal*

The top portion of a Lurgi gasifier acts as a distillation section in which large quantities of tar and tar oil are produced. These products are carried from the gasifier with the raw gas and are subsequently condensed from the gas. A Lurgi designed tar-oil separation unit will be utilized to separate tar and oil from the waste water. A schematic diagram of this system is shown in figure 6. The separators utilize the specific gravity difference between tar, oil, and water to effect a separation. Tar is defined as being all organics heavier than water, and oil is defined as being all organics lighter than water. The expected quantities of tar and tar oil to be produced in the Burnham Complex also are shown in figure 6.

As illustrated in figure 6, there will be two tar-oil separation systems, one of which will handle the high solids (high halogen) condensate, and the other will handle the "clean" condensate (low solids). After tar-oil separation, each of these condensate streams will be sent to the Phenosolvan area, a proprietary Lurgi process for the extraction of phenols and removal of acid gases.

### *Phenol Removal*

In the Phenosolvan process, isopropyl ether is used as a solvent to extract phenols and other organic compounds from the waste water. As illustrated in figure 7, Lurgi guarantees a maximum of 20 ppm of steam volatile phenols in the effluent water.

Separate extractors will be utilized for the high solids and low solids condensate streams.

### *Ammonia and Acid Gas Removal*

Before the waste water can be used as cooling tower makeup, the acid gases ($CO_2$ and $H_2S$) and ammonia must be stripped out of the water. In the Burnham Complex, the acid gases will be stripped from the water in a reboiled, pressurized column. The overhead from this deacidifier has the composition shown in table 5. The gas is sent to the low pressure Stretford absorber, where 99.9 percent of the $H_2S$ is converted to elemental suflur.

Following acid gas removal, the ammonia will be stripped from the waste water, and the stripped gases will be condensed as an aqueous ammonia solution. About 250 tons per day of anhydrous ammonia could be produced from this stream. The water from the bottom of the ammonia stripper will be used as cooling tower makeup water. El Paso has simulated this water composition at the SASOL plant and has been studying the operability of a test cooling tower and heat exchanger for about one year. The results of these tests are being used to study the characteristics

FIGURE 7

PHENOL EXTRACTION SECTION

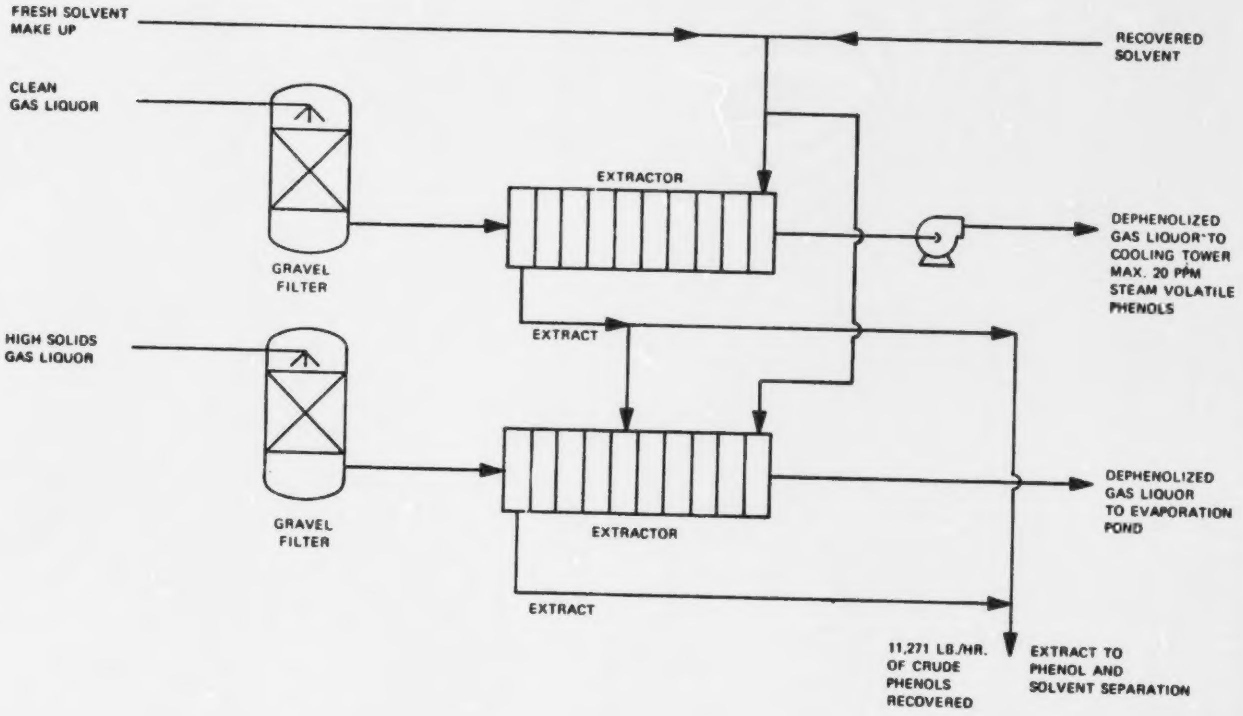

FRESH SOLVENT MAKE UP
RECOVERED SOLVENT
CLEAN GAS LIQUOR
GRAVEL FILTER
EXTRACTOR
DEPHENOLIZED GAS LIQUOR TO COOLING TOWER MAX. 20 PPM STEAM VOLATILE PHENOLS
EXTRACT
HIGH SOLIDS GAS LIQUOR
GRAVEL FILTER
EXTRACTOR
DEPHENOLIZED GAS LIQUOR TO EVAPORATION POND
EXTRACT
11,271 LB./HR. OF CRUDE PHENOLS RECOVERED
EXTRACT TO PHENOL AND SOLVENT SEPARATION

Table 5. Composition of acid gases stripped from gas liquor

| Component | Mol % (dry) |
|---|---|
| $CO_2$ | 95.91 |
| $H_2S$ | 4.09 |
| Total | 100.00 |

Total dry gas = 8,853 lb mols/hr

of the Burnham cooling water system, such as allowable cycles of concentration, foaming tendency, slime buildup in tower, and heat transfer characteristics of the test heat exchanger.

## SUMMARY

El Paso Natural Gas Company has designed the Burnham Coal Gasification Complex with full consideration to minimizing air and water pollutant emissions. Sulfur emissions in the high-Btu gas production areas will be minimized by utilizing the Stretford Process to remove the $H_2S$ from the acid gas from the Rectisol plant. Lurgi gasifiers will be used to produce a fuel gas, and a high pressure Stretford unit will be used to effectively remove the $H_2S$ from this fuel gas. This fuel gas system will provide the fuel for steam and power production. A coal-fired boiler-stack gas scrubbing system will not be utilized. Particulate emissions from the Burnham Complex will be low, since 1½ inch X ¼ inch coal is typically fed to Lurgi gasifiers. The prevailing $NO_x$ regulations will be easily achieved with the planned low-Btu gas power and steam generating systems.

Water pollutants will be minimized through the use of proprietary Lurgi processes for the removal of tar, tar oil, phenols, and acid gases from the gas liquor (dirty condensate). Water reuse will be maximized by utilizing much of the gas liquor as cooling tower makeup water.

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# ENVIRONMENTAL ASPECTS OF THE WESCO COAL GASIFICATION PLANT

**Thomas E. Berty and James M. Moe***

*Abstract*

*Some of the essential processing factors in the Lurgi gasification process are very briefly reviewed. The quantity and types of byproducts are described, and the environmentally related problems are presented. The efforts to conserve water are discussed, and the manner in which the air quality standards are achieved is described in detail.*

## INTRODUCTION

The present and projected shortage of natural gas has led to several ambitious projects to produce substitute natural gas from coal. At the present time, four projects to produce SNG from coal by the Lurgi pressure gasification process are in various phases of engineering design. Each of these plants will produce 250 million of SNG. It seems likely that more plants utilizing the Lurgi process will be planned for the future.

Any process based on coal will inevitably encounter many environmental problems, and the Lurgi process is no exception. A large proportion of the total plant cost is devoted to environmentally related matters and to byproduct treating.

### *Process Descriptions,*

Figure 1 shows, in block form, the overall processing sequence. In the gasification section, coal is reacted with oxygen and steam to form a crude gas which is primarily a mixture of carbon dioxide, carbon monoxide, hydrogen, methane, and water vapor. The gas stream also contains ammonia, hydrogen cyanide, hydrogen sulfide, carbonyl sulfide, and carbon disulfide. Halogens in the coal will form their respective acid. Additional compounds in the crude gas are the hydrocarbon byproducts which consists of tar, tar-oil, naphtha, crude phenols, and fatty acids.

The function of the other processing units in figure 1 is to upgrade the crude gas stream, and separate and recover the various byproducts. All of the tars, tar-oil, naphtha, phenols, and fatty acids are separated from the gas stream by cooling. This also condenses most of the water from the gas. This aqueous mixture (called gas-liquor) is processed by the Phenosolvan unit and the gas-liquor unit.

The partially purified gas stream, after cooling, is treated in the Rectisol plant to remove acid gas constituents: $CO_2$, $H_2S$, COS, $CS_2$, and HCN. After this treatment, the gas is a very pure mixture of CO, $CO_2$, $H_2$, and $CH_4$ which is then upgraded to pipeline quality SNG by the methanation unit.

### *Auxiliary Units*

The auxiliary units include a cryongenic air separation plant to produce oxygen, a Claus sulfur plant for sulfur recovery, a coal-fired steam plant with flue gas treating for $SO_2$ removal, cooling water systems, byproduct storage and loading facilities, safety systems, effluent water treatment and reuse systems, ash handling systems, and numerous service facilities and buildings.

### *Water Usage*

The WESCO plant gasifies 21,800 tons per day of coal and requires 5,100 gpm of raw water intake. This amounts to 1.4 pounds of water per pound of coal. This includes water required by the coal mining operation as well as the gasification plant and its auxiliary utility services.

Since the supply of water to the project is contractually limited, every effort has been made in the design to conserve water usage and to maximize the recycle and reuse. Some of the major design features used to achieve those objectives are:

1. About 250,000 hp of large compressor-driving steam turbines will be provided with air-cooled exhaust steam condensers. The condensed steam will be recovered and reused as boiler feedwater. The air-cooled condensers will provide 2 billion Btu/hour of heat removal. If cooling water were used for this heat removal, the evaporative water loss from the cooling tower would be increased by about 4,000 gpm (6,400 acre-ft per year). This would almost double the raw water feed requirement. The larger cooling tower would also incur larger windage losses.

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*Principal Process Engineers, Fluor Engineers and Constructors, Inc., Los Angeles, California.

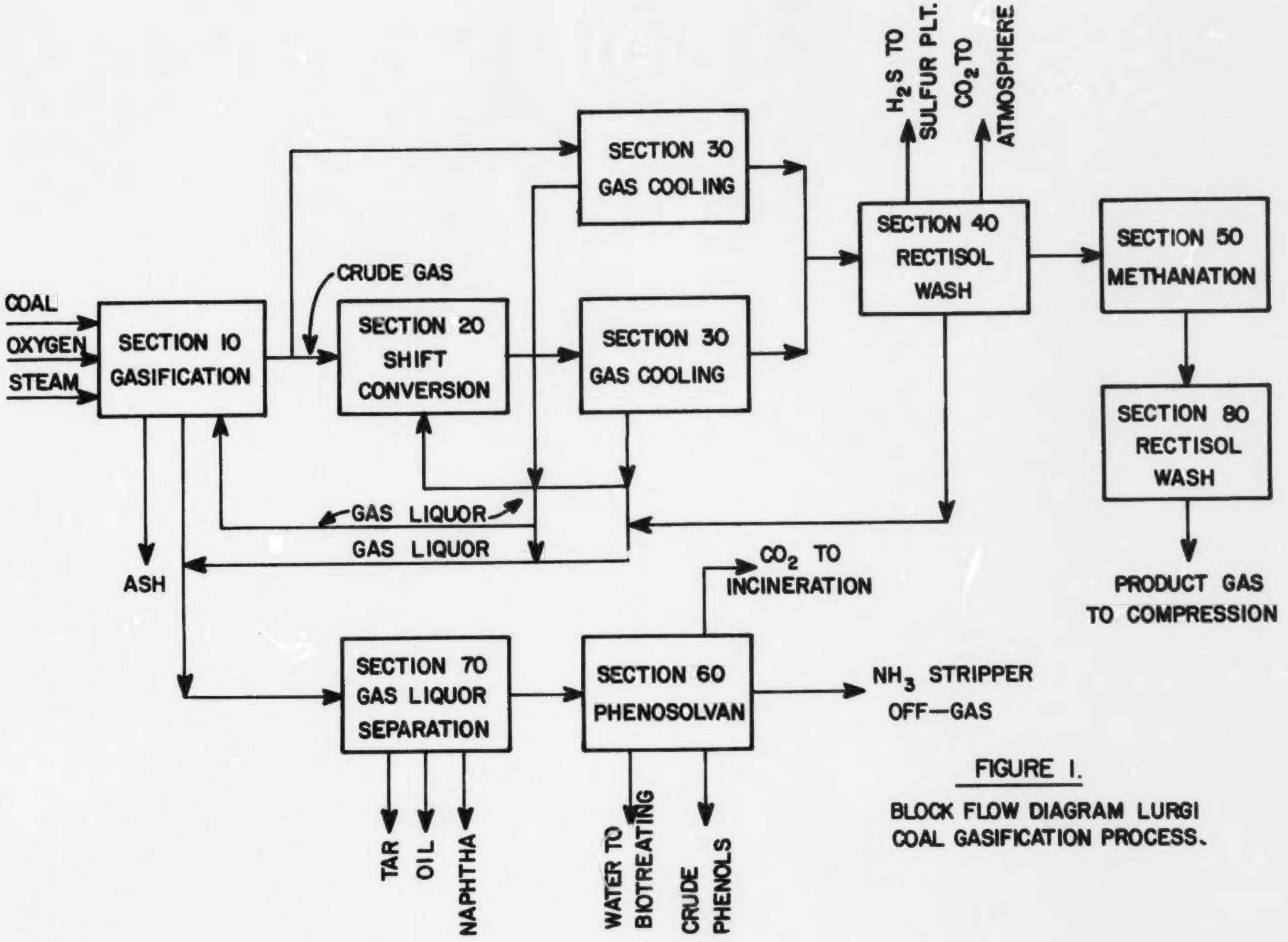

COAL
OXYGEN
STEAM
SECTION 10 GASIFICATION
ASH
CRUDE GAS
SECTION 20 SHIFT CONVERSION
SECTION 30 GAS COOLING
SECTION 30 GAS COOLING
GAS LIQUOR
GAS LIQUOR
SECTION 40 RECTISOL WASH
H2S TO SULFUR PLT.
CO2 TO ATMOSPHERE
SECTION 50 METHANATION
SECTION 80 RECTISOL WASH
PRODUCT GAS TO COMPRESSION
CO2 TO INCINERATION
SECTION 70 GAS LIQUOR SEPARATION
TAR
OIL
NAPHTHA
SECTION 60 PHENOSOLVAN
NH3 STRIPPER OFF—GAS
WATER TO BIOTREATING
CRUDE PHENOLS

FIGURE 1.

BLOCK FLOW DIAGRAM LURGI COAL GASIFICATION PROCESS.

2. Raw water will be treated to boiler feed water quality, and then it is converted to high-pressure steam. A major portion of this steam is fed into the gasifiers. The unreacted steam is recovered as a "gas liquor" from which byproduct hydrocarbons that include phenols will be recovered. The dephenolized gas liquor is stripped of dissolved ammonia and hydrogen sulfide, then it is treated for removal of oil and suspended solids, and finally it is biochemically oxidized in two stages of biotreating. The treated and clarified effluent will then be reused as water makeup to the plant cooling water system. In fact, it will supply 100 percent of the cooling water makeup needs.

3. Byproduct water produced in the methanation reaction will be recovered and reused as boiler feedwater.

4. Mechanical refrigeration will provide the low temperatures needed in the Rectisol Unit. Mechanical refrigeration with air-cooled condensers was chosen in preference to an absorption refrigeration system to avoid the very large cooling water evaporation losses associated with absorption refrigeration.

5. Water will be extracted from water treatment sludges and recycled for reuse.

6. Blowdown from the cooling water system will be reused for quenching of the ash from both the gasifiers and the coal-fired boilers.

The plant will require only 8,200 acre-ft per year, or about 75 percent of the contract water availability. This plan provides a margin of safety for any unforeseen contingencies.

Table 1 shows the Water Requirements and Disposition for the project. The ultimate disposition of the 5,100 gpm intake water can be briefly summarized as:

| | |
|---|---|
| Process consumption | 10.2% |
| Return to atmosphere | 69.6% |
| Disposal to mine reclamation | 8.4% |
| Others | 11.8% |
| | 100.0% |

Because a coal-fired boiler is used, particulate emissions must also be controlled. The majority of the particulates are removed with electrostatic precipitators. However, the stack-gas treating process also removes some particulates, so the net result is that only 1.8 tons/day of particulate matter is discharged to the atmosphere.

The tail gas from the Claus sulfur plant is incinerated in the boiler (to oxidize the $H_2S$ to $SO_2$) and then treated with the flue gas for $SO_2$ removal. A decision has not yet been made on the stack-gas treating process, although the Chiyoda 101 and the MKK-Wellman Lord processes are being considered. The MKK process recovers the sulfur as sulfuric acid while the Chiyoda process yields gypsum.

The $CO_2$ stream vented to the atmosphere in the Rectisol plant amounts to 284 million scfd. This stream is 98.3 percent $CO_2$, with the balance being hydrocarbons ($CH_4$, $C_2H_6$, and $C_2H_4$). About 35 ppm of COS is also in the gas. The presence of ethylene in this stream, even though it amounts to only 0.35 volume %, may be somewhat of a problem.

In some locations, it may be possible to vent this material directly to the air. In situations where this is not permitted, it may be necessary to install another cleanup device such as a catalytic oxidizer.

Table 1. Water Requirements and Disposition

| | GPM | % |
|---|---|---|
| Process Consumption | | |
| To supply hydrogen | 1,120 | |
| Produced as methanation by product | 600 | |
| Net Consumption | 520 | 10.2 |
| Return to Atmosphere | | |
| Evaporation: | | |
| From raw water ponds | 420 | |
| From cooling tower | 1,760 | |
| From quenching hot ash | 150 | |
| From pelletizing sulfur | 250 | |
| From wetting of mine roads | 730 | |
| | 3,310 | |
| Via stack gases[1]: | | |
| From steam blowing of boiler tubes | 200 | |
| From stack gas $SO_2$ scrubbers | 40 | |
| | 240 | |
| Total return to atmosphere | 3,550 | 69.6 |
| Disposal to Mine Reclamation | | |
| In water treating sludges | 100 | |
| In wetted boiler ash | 30 | |
| In wetted gasifier ash | 300 | |
| Total disposal to mine | 430 | 8.4 |
| Others | | |
| Retained in slurry pond | 20 | |
| Miscellaneous mine uses | 580 | |
| Total others | 600 | 11.8 |
| GRAND TOTAL | 5,100 | 100.0 |

---

(1) Does not include water derived from burning of boiler fuel.

The stream superheater furnace effluent meets environmental regulations without treatment by firing a low-sulfur fuel oil.

The air emissions described are those which occur during normal operation of the plant. Some intermittent emissions also occur during regeneration of the shift catalyst and certain startup operations. These emissions are handled by incinerating them in the boiler plant, after which they are treated by the stack-gas treating process.

### *Solid Waste*

Waste solids are disposed of by being buried in the mine. These materials consist of ash from the boiler plant and from the gasifiers, coal wash plant waste slurry, and the combined sludge from the water clarifier, biotreating unit, and the sanitary waste treatment.

### *Conclusion*

It can be appreciated that a great deal of effort and money is required to meet the environmental regulations in a coal gasification plant. Nevertheless, meeting present day regulations is within the realm of proven technology, and, at the same time, they allow the designer some latitude of choice in selecting appropriate processes. For example, in the WESCO plant the decision was made to use a coal-fired boiler and stack-gas treating for $SO_2$ removal rather than one of the so-called "clean fuel cycles". This choice was made on the basis of the specific economic factors appropriate to the WESCO plant.

It is important to note that about 70 percent of the water will be returned to the regional atmospheric environment and will eventually become rainwater. About 20 percent of the water is returned to the environment in the form of liquid water.

It is also important to note that the 20 percent will be disposed of on-site, principally as sludges and wetted ash used in the reclamation of the coal mining area.

A schematic diagram of the water treatment and reuse systems is included as figure 2 and graphically illustrates the extent to which recycle and water reuse has been designed into the plant:

(1) As already discussed, the water used to provide turbine steam is condensed and recycled for 100 percent reuse.

(2) The water fed into the gasifiers (as steam) provides the hydrogen required to convert the coal to methane (SNG). After serving as a gasification reactant, the excess steam is condensed as phenolic gas liquor from which useful byproduct phenols are then extracted. After oil and solids removal, and biotreatment, the recovered water provides 100 percent of the makeup needs for the process cooling water system.

This water has now served three useful functions:

(a) Supplied necessary reactant in the conversion of coal to SNG;

(b) Served as the medium for removal and recovery of byproduct phenols (as well as ammonia);

(c) Supplied 100 percent of the cooling water makeup needs.

(3) The cooling water system is a closed loop with an evaporative cooling tower. Water is recycled and reused in the system about 3-6 times before the buildup of dissolved salts necessitates a blowdown to maintain a tolerable level of salts within the circulating water. Even this blowdown is reused; it quenches the hot ashes from the gasifiers.

(4) Boiler blowdown water, rinse waters from the water demineralizers, treated effluent from the sanitary system biotreatment unit, and excess process condensate will be selectively reused to provide:

(a) Water for wetting of roads in the mining area (dust abatement);

(b) Water for pelletizing the byproduct sulfur;

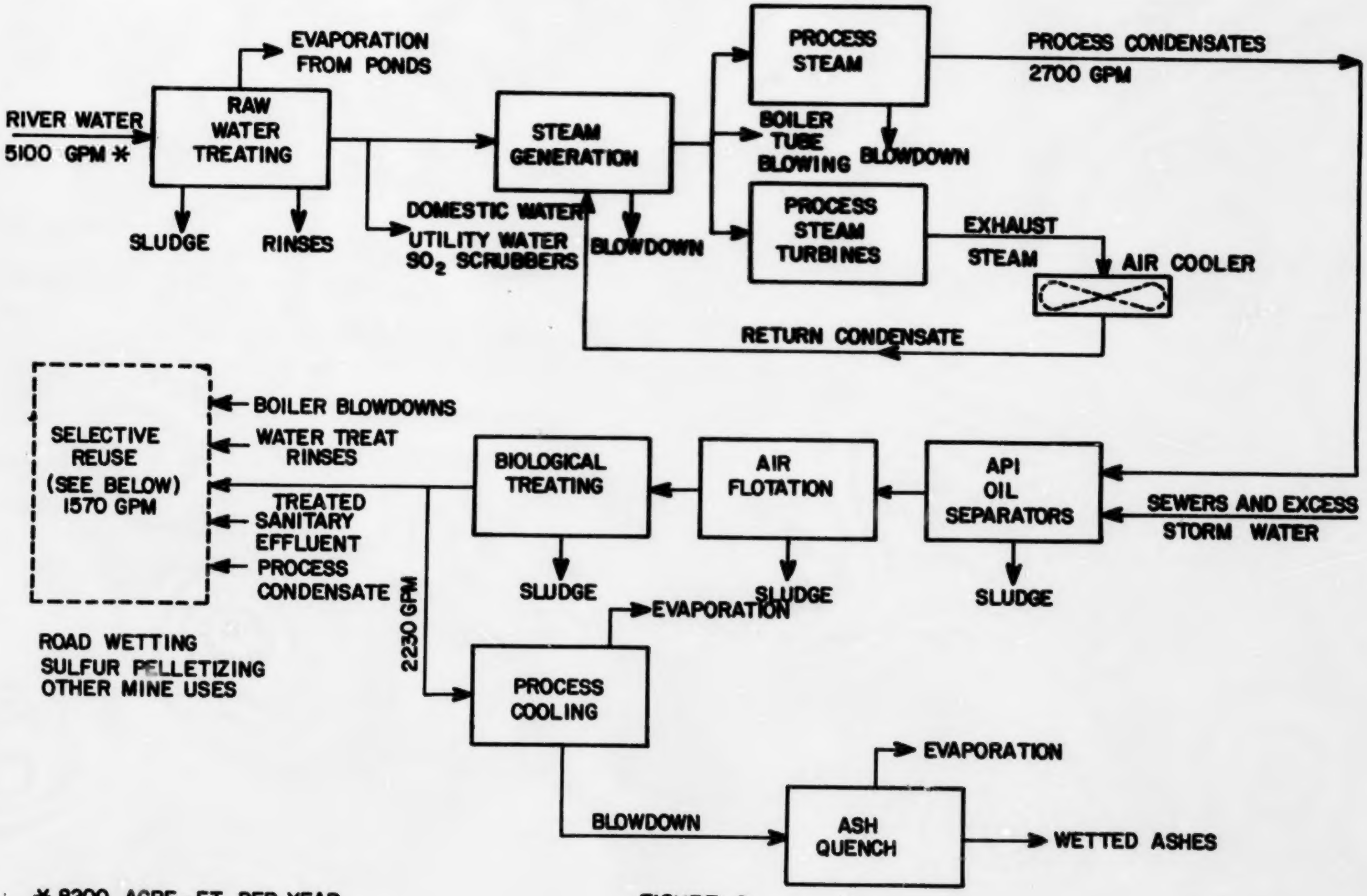
(c) Other uses in the mining and coal processing operations.

### *Air Emissions*

All of the materials emitted to the atmosphere occur at three sources: the boiler plant stack, the steam superheater furnace, and the $CO_2$ stream discharged from the Rectisol plant. Figure 3 shows the arrangement and the overall sulfur balance.

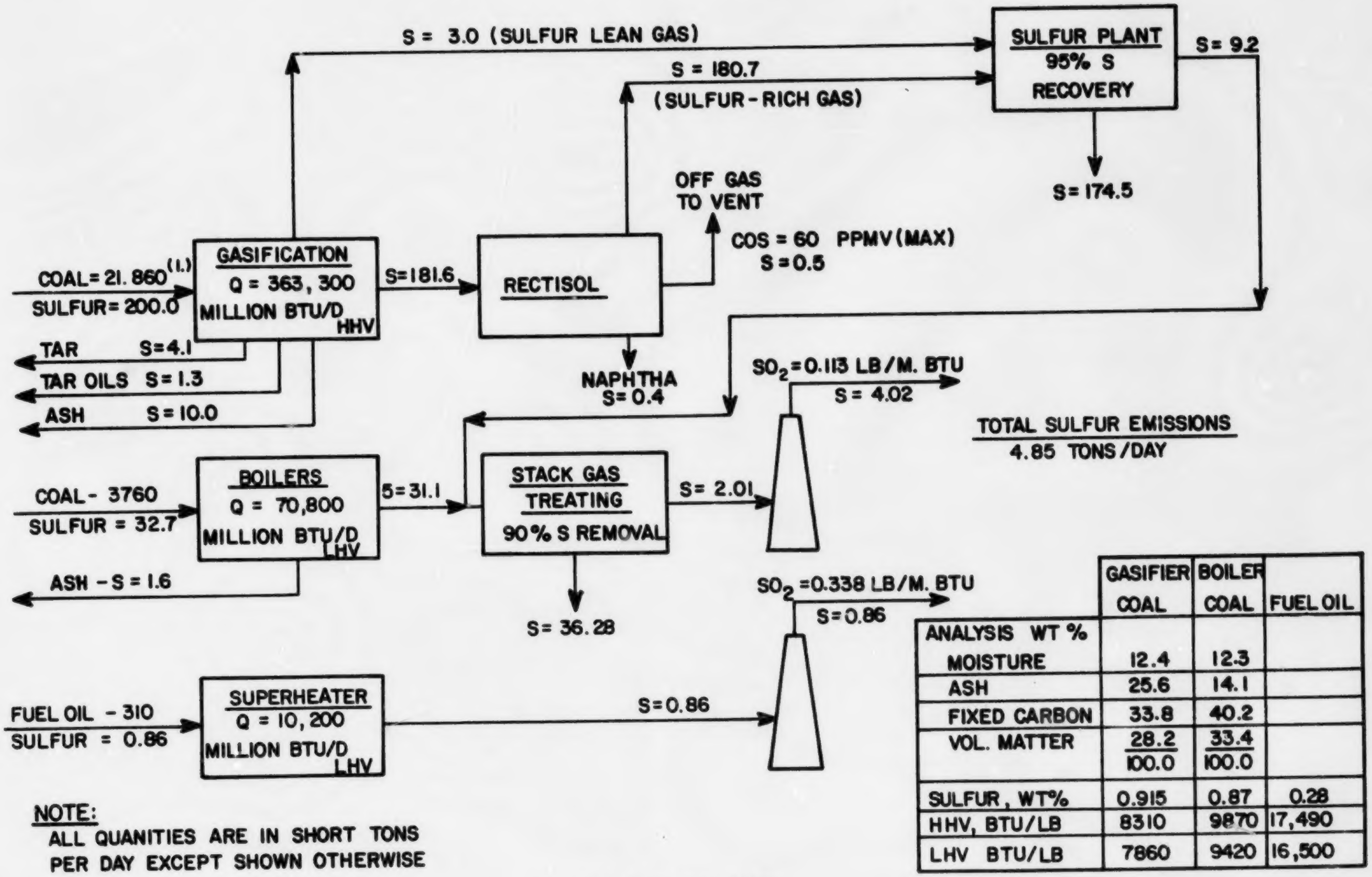
If the allowable $SO_2$ emissions were decreased by say, a factor of two, this choice of processes would not be possible. The point we wish to make is that the current environmental regulations are taxing technology to the utmost, and more stringent regulations would force matters into unproven techniques of highly uneconomical alternatives.

## WATER TREATMENT AND REUSE SYSTEMS


EVAPORATION FROM PONDS
RIVER WATER
5100 GPM *
RAW WATER TREATING
SLUDGE
RINSES
DOMESTIC WATER
UTILITY WATER
$SO_2$ SCRUBBERS
STEAM GENERATION
BLOWDOWN
PROCESS STEAM
BOILER TUBE BLOWING
BLOWDOWN
PROCESS STEAM TURBINES
EXHAUST STEAM
AIR COOLER
RETURN CONDENSATE
PROCESS CONDENSATES
2700 GPM
SELECTIVE REUSE (SEE BELOW) 1570 GPM
BOILER BLOWDOWNS
WATER TREAT RINSES
TREATED SANITARY EFFLUENT
PROCESS CONDENSATE
ROAD WETTING
SULFUR PELLETIZING
OTHER MINE USES
BIOLOGICAL TREATING
SLUDGE
AIR FLOTATION
SLUDGE
API OIL SEPARATORS
SLUDGE
SEWERS AND EXCESS STORM WATER
2230 GPM
EVAPORATION
PROCESS COOLING
BLOWDOWN
ASH QUENCH
EVAPORATION
WETTED ASHES

* 8200 ACRE-FT. PER YEAR

FIGURE 2


S = 3.0 (SULFUR LEAN GAS)
SULFUR PLANT
95% S
RECOVERY
S= 9.2
S = 180.7
(SULFUR - RICH GAS)
S= 174.5
OFF GAS
TO VENT
COS = 60 PPMV(MAX)
S = 0.5
COAL = 21.860 (1.)
SULFUR = 200.0
GASIFICATION
Q = 363,300
MILLION BTU/D
HHV
S=181.6
RECTISOL
TAR S=4.1
TAR OILS S = 1.3
ASH S = 10.0
NAPHTHA
S = 0.4
$SO_2$ = 0.113 LB/M. BTU
S = 4.02
TOTAL SULFUR EMISSIONS
4.85 TONS/DAY
COAL - 3760
SULFUR = 32.7
BOILERS
Q = 70,800
MILLION BTU/D
LHV
5=31.1
STACK GAS
TREATING
90% S REMOVAL
S= 2.01
ASH - S = 1.6
S= 36.28
$SO_2$ = 0.338 LB/M. BTU
S=0.86
FUEL OIL - 310
SULFUR = 0.86
SUPERHEATER
Q = 10,200
MILLION BTU/D
LHV
S=0.86

| | GASIFIER COAL | BOILER COAL | FUEL OIL |
|---|---|---|---|
| ANALYSIS WT % | | | |
| MOISTURE | 12.4 | 12.3 | |
| ASH | 25.6 | 14.1 | |
| FIXED CARBON | 33.8 | 40.2 | |
| VOL. MATTER | 28.2 | 33.4 | |
| | 100.0 | 100.0 | |
| SULFUR, WT% | 0.915 | 0.87 | 0.28 |
| HHV, BTU/LB | 8310 | 9870 | 17,490 |
| LHV BTU/LB | 7860 | 9420 | 16,500 |

NOTE:
ALL QUANITIES ARE IN SHORT TONS PER DAY EXCEPT SHOWN OTHERWISE

SULFUR BALANCE
FIGURE 3

(1.) DESIGN VALUE BASED ON LURGI'S COMPOSITE SAMPLES

# ANALYSES OF TARS, CHARS, GASES, AND WATER FOUND IN EFFLUENTS FROM THE SYNTHANE PROCESS*

**Albert J. Forney, William P. Haynes, Stanley J. Gasior,
Glenn E. Johnson, and Joseph P. Strakey, Jr.†**

*Abstract*

*Extensive studies have been made of the various effluents found in the Synthane coal-to-gas process. Analyses have been made of the waters, gases, and trace elements present in some of the streams. Results of analyses show the water effluents are the area where extensive research is needed.*

## INTRODUCTION

One of the answers to the shortage of natural gas is to convert coal to a high-Btu gas. Four coal-conversion projects are proceeding to the prototype plant scale (75 to 120 tpd coal utilization) (ref. 1). An aspect of coal gasification that is of major importance is the possible pollution resulting from the process. While the four prototype plants have different types of processing units, they will have similar effluents. This paper discusses this aspect of the Synthane process, based on the Bureau of Mines laboratory-scale gasifier at Bruceton, Pa.

The advantage of the Bruceton laboratory-scale Synthane gasifier in studying effluent problems is that the waters, tars, gases, and solids streams are representative of those that will be obtained from a commercial operation. There will be some difference due to temperatures and variations in steam-oxygen feed quantities, but the samples of streams discussed in the following tables will be quite representative of larger scale operation. This will also be true of the byproducts of the methanation step. Most of our knowledge of these effluents is based on the work at the Bruceton laboratories of the Pittsburgh Energy Research Center (PERC).

## ACKNOWLEDGMENT

At PERC the solids, water, and tar analyses were mostly performed by the General Analysis group headed by H. Schultz, with special thanks due to F. E. Walker, J. F. Smith, and M. F. Ferrer. Other water, tar, and gas anlyses were made by the Spectro-Physics group headed by R. A. Friedel with A. G. Sharkey and C. E. Schmidt. Trace element analysis (table 2) of the waters was done by Charles E. Taylor of EPA, and the tar and gas were analyzed by Bernard Keisch of Carnegie-Mellon University. The HCN analyses were performed by Dr. Schultz's group.

## THE OVERALL PROCESS

The overall process is shown schematically in figure 1. It shows the 75-tpd pilot plant which was designed by the Lummus Co. and is being built as Bruceton, Pa., by the Rust Engineering Co. The major units shown are the gasifier, shift converter, purification systems, and methanators. Each of these units has its byproduct streams.

## WATER ANALYSIS

The major effluent problem is the contaminated condensate from the gasifier. The Bruceton laboratory gasifier, shown in figure 2, condenses the water, tars, and dusts in two water-cooled condensers operated at 100° and 50°C. Table 1 shows the analysis of the condensate from gasification tests with a number of different coals compared with a coke-plant weak ammonia liquor. Bethlehem Steel Co. at its Bethlehem, Pa., plant (ref. 2) has reduced the phenol level of its weak ammonia liquor to 100 ppb by biological oxidation and has reduced the thiocyanates by an average of 70 percent. This plant has been operating at Bethlehem for over 10 years, and the effluent of the plant satisfactorily meets Pennsylvania pollution requirements. Therefore, we consider this system a satisfactory means of solving the effluent problems of the Synthane plant. However, work is continuing on new and better methods of alleviating these problems.

Additional analyses of the condensate were performed by the Environmental Protection Agency at its Southeast Environmental Research Laboratory; the trace elements in the water are shown in table 2.

For a commercial coal-to-gas plant, this water would be purified as completely as possible and then

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*Bureau of Mines Application of Improved Technology to Provide Clean Energy Program, Technical Progress Report 76, U.S. Department of the Interior, January 1974.

†All are with the Pittsburgh Energy Research Center, Pittsburgh, Pa: Forney is research supervisor, Haynes and Strakey are supervisory chemical engineers, Gasior and Johnson are chemical engineers.

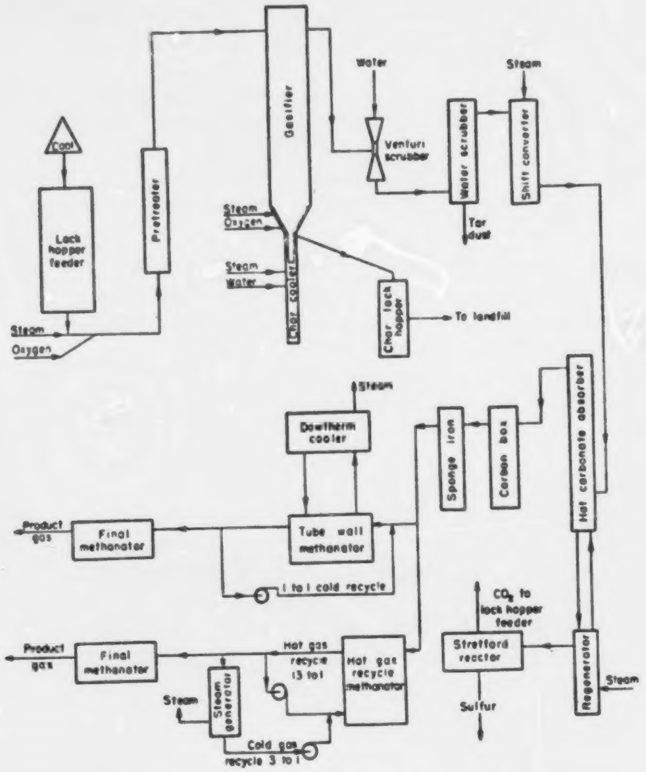
Coal
Lock hopper feeder
Steam
Oxygen
Pretreater
Gasifier
Steam
Oxygen
Steam
Water
Char cooler
Char lock hopper
To landfill
Water
Venturi scrubber
Water scrubber
Tar dust
Steam
Shift converter
Steam
Dowtherm cooler
Product gas
Final methanator
Tube wall methanator
1 to 1 cold recycle
Sponge iron
Carbon box
Hot carbonate absorber
$CO_2$ to lock hopper feeder
Stretford reactor
Sulfur
Regenerator
Steam
Product gas
Final methanator
Hot gas recycle 13 to 1
Hot gas recycle methanator
Steam
Steam generator
Cold gas recycle 3 to 1

FIGURE 1. - Flowsheet of prototype Synthane process.

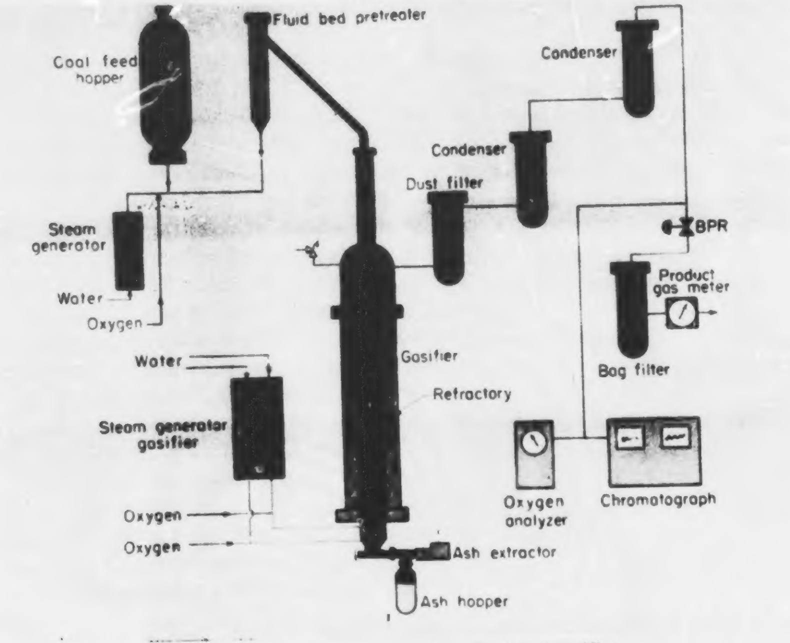
Fluid bed pretreater
Coal feed hopper
Condenser
Condenser
Dust filter
Steam generator
Water
Oxygen
BPR
Product gas meter
Water
Gasifier
Bag filter
Refractory
Steam generator gasifier
Oxygen
Oxygen
Oxygen analyzer
Chromatograph
Ash extractor
Ash hopper

Figure 2. Forty-atmosphere fluid-bed gasifier.

Table 1. Byproduct water analysis, from Synthane gasification of various coals, mg/l (except pH)

| | Coke plant | Illinois No. 6 coal | Wyoming sub-bitu-minous coal | Illi-nois lignite | North Dakota lignite | Western Kentucky coal | Pitts-burgh seam coal |
|---|---|---|---|---|---|---|---|
| pH | 9 | 8.6 | 8.7 | 7.9 | 9.2 | 8.9 | 9.3 |
| Suspended solids | 50 | 600 | 140 | 24 | 64 | 55 | 23 |
| Phenol | 2,000 | 2,600 | 6,000 | 200 | 6,600 | 3,700 | 1,700 |
| COD | 7,000 | 15,000 | 43,000 | 1,700 | 38,000 | 19,000 | 19,000 |
| Thiocyanate | 1,000 | 152 | 23 | 21 | 22 | 200 | 188 |
| Cyanide | 100 | 0.6 | 0.23 | 0.1 | 0.1 | 0.5 | 0.6 |
| $NH_3$ | 5,000 | 8,100* | 9,520 | 2,500 | 7,200 | 10,000 | 11,000 |
| Chloride | - | 500 | - | 31 | - | - | - |
| Carbonate | - | 6,000† | - | - | - | - | - |
| Bicarbonate | - | 11,000† | - | - | - | - | - |
| Total sulfur | - | 1,400‡ | - | - | - | - | - |

* 85 percent free $NH_3$.

† Not from same analysis.

‡ $S^{=}$ = 400; $SO_3^{=}$ = 300; $SO_4^{=}$ = 1,400; $S_2O_3^{=}$ = 1,000.

Table 2. Trace elements in condensate from an Illinois No. 6 coal gasification test

| | No. 1 | No. 2 | Average (by weight) |
|---|---|---|---|
| ppm: | | | |
| Calcium | 4.4 | 3.6 | 4 |
| Iron | 2.6 | 2.9 | 3 |
| Magnesium | 1.5 | 1.8 | 2 |
| Aluminum | 0.8 | 0.7 | 0.8 |
| ppb: | | | |
| Selenium | 401 | 323 | 360 |
| Potassium | 117 | 204 | 160 |
| Barium | 109 | 155 | 130 |
| Phosphorus | 82 | 92 | 90 |
| Zinc | 44 | 83 | 60 |
| Manganese | 36 | 38 | 40 |
| Germanium | 32 | 61 | 40 |
| Arsenic | 44 | 28 | 30 |
| Nickel | 23 | 34 | 30 |
| Strontium | 33 | 24 | 30 |
| Tin | 25 | 26 | 20 |
| Copper | 16 | 20 | 20 |
| Columbium | 7 | 5 | 6 |
| Chromium | 4 | 8 | 6 |
| Vanadium | 4 | 2 | 3 |
| Cobalt | 1 | 2 | 2 |

Table 3. Components in gasifier gas, ppm

| | Illinois No. 6 coal | Illinois char | Wyoming subbituminous coal | Western Kentucky coal | North Dakota lignite | Pittsburgh seam coal |
|---|---|---|---|---|---|---|
| $H_2S$................ | 9,800 | 186 | 2,480 | 2,530 | 1,750 | 860 |
| COS................ | 150 | 2 | 32 | 119 | 65 | 11 |
| Thiophene........... | 31 | 0.4 | 10 | 5 | 13 | 42 |
| Methyl thiophene.... | 10 | 0.4 | - | - | - | 7 |
| Dimethyl thiophene.. | 10 | 0.5 | - | - | 11 | 6 |
| Benzene............. | 340 | 10 | 434 | 100 | 1,727 | 1,050 |
| Toluene............. | 94 | 3 | 59 | 22 | 167 | 185 |
| $C_8$ aromatics........ | 24 | 2 | 27 | 4 | 73 | 27 |
| $SO_2$................ | 10 | 1 | 6 | 2 | 10 | 10 |
| $CS_2$................ | 10 | - | - | - | - | - |
| Methyl mercaptan.... | 60 | 0.1 | 0.4 | 33 | 10 | 8 |

Table 4. Mass spectrometric analyses of the benzene-soluble tar, volume-percent

| Structural type (includes alkyl derivatives) | Run HP-1 No. 92, Illinois[1] No. 6 coal | Run HPL No. 94, lignite | Run HPM No. 11 Montana subbituminous coal | Run HP-118 No. 118,[1] Pittsburgh seam coal |
|---|---|---|---|---|
| Benzenes................. | 2.1 | 4.1 | 3.9 | 1.9 |
| Indenes.................. | [2]8.6 | 1.5 | 2.6 | [2]6.1 |
| Indans................... | 1.9 | 3.5 | 4.9 | 2.1 |
| Naphthalenes............. | 11.6 | 19.0 | 15.3 | 16.5 |
| Flourenes................ | 9.6 | 7.2 | 9.7 | 10.7 |
| Acenaphthenes............ | 13.5 | 12.0 | 11.1 | 15.8 |
| 3-ring aromatics......... | 13.8 | 10.5 | 9.0 | 14.3 |
| Phenylnaphthalenes....... | 9.8 | 3.5 | 6.4 | 7.6 |
| 4-ring pericondensed..... | 7.2 | 3.5 | 4.9 | 7.6 |
| 4-ring catacondensed..... | 4.0 | 1.4 | 3.0 | 4.1 |
| Phenols.................. | 2.8 | 13.7 | 5.5 | 3.0 |
| Naphthols................ | ([2]) | 9.7 | 9.6 | ([2]) |
| Indanols................. | 0.9 | 1.7 | 1.5 | 0.7 |
| Acenaphthenols........... | - | 2.5 | 4.6 | 2.0 |
| Phenanthrols............. | 2.7 | - | 0.9 | - |
| Dibenzofurans............ | 6.3 | 5.2 | 5.6 | 4.7 |
| Dibenzothiophenes........ | 3.5 | 1.0 | 1.5 | 2.4 |
| Benzonaphthothiophenes... | 1.7 | - | - | - |
| N-heterocyclics[3]......... | (10.8) | (3.8) | (5.3) | (8.8) |
| Average molecular weight. | 212 | 173 | 230 | 202 |

[1] Spectra indicate traces of 5-ring aromatics.

[2] Includes any naphthol present (not resolved in these spectra).

[3] Data on N-free basis since isotope corrections were estimated.

Table 5. Sulfur content of coals and products, weight-percent (except gas)

| | Coal type | | | |
|---|---|---|---|---|
| | Pittsburgh seam coal | Illinois No. 6 coal | Montana subbituminous coal | North Dakota lignite |
| Coal | 1.5 | 3.5 | 0.6 | 1.1 |
| Char | 0.5 | 1.1 | 0.4 | 0.6 |
| Tar | 0.8 | 2.7 | 0.5 | 1.0 |
| Condensate | <0.1 | <0.1 | <0.1 | <0.1 |
| Gas (vol-pct) | 0.3 | 1.1 | 0.4 | 0.3 |

Table 6. Representative analyses of coals and chars, weight-percent

| | Illinois No. 6 coal | Western Kentucky coal | Wyoming subbituminous coal | North Dakota lignite | Pittsburgh seam coal |
|---|---|---|---|---|---|
| Coals: | | | | | |
| Moisture | 8.3 | 4.3 | 18.1 | 20.6 | 2.5 |
| Volatile matter | 37.5 | 34.6 | 31.9 | 32.9 | 30.9 |
| Fixed carbon | 43.0 | 44.5 | 32.0 | 38.2 | 51.5 |
| Ash | 11.2 | 16.6 | 18.0 | 8.3 | 15.1 |
| Hydrogen | 5.3 | 4.7 | 5.4 | 5.7 | 4.7 |
| Oxygen | 15.9 | 10.9 | 30.3 | 32.6 | 9.3 |
| Carbon | 63.0 | 62.7 | 45.2 | 51.5 | 68.4 |
| Nitrogen | 1.1 | 1.2 | 0.6 | 0.7 | 1.2 |
| Sulfur | 3.5 | 3.9 | 0.5 | 1.2 | 1.3 |
| Chars (from above coals): | | | | | |
| Moisture | 0.8 | 1.2 | 0.5 | 1.2 | 1.4 |
| Volatile matter | 4.0 | 4.8 | 5.1 | 10.0 | 1.6 |
| Fixed carbon | 69.9 | 63.3 | 38.1 | 50.2 | 69.3 |
| Ash | 25.3 | 30.7 | 56.3 | 38.6 | 27.7 |
| Hydrogen | 1.0 | 1.0 | 1.0 | 0.9 | 1.0 |
| Oxygen | 1.3 | 1.1 | 1.2 | 0.0 | 1.7 |
| Carbon | 70.4 | 64.5 | 40.6 | 58.9 | 68.9 |
| Nitrogen | 0.6 | 0.7 | 0.4 | 0.2 | 0.5 |
| Sulfur | 1.4 | 2.0 | 0.5 | 2.0 | 0.2 |

used as a recycled cooling water; therefore, it could not possibly contaminate any streams.

## GAS ANALYSIS

Besides the large quantities of $H_2$, CO, $CO_2$, $CH_4$, and $C_2H_6$ made in the gasifier, a number of trace components are found. These are shown in table 3 which shows the sulfur compounds plus the BTX (benzene-toluene-xylene) components. The sulfur compounds must be removed before methanation because of their poisoning effect on the nickel catalyst. The use of Benfield* hot-carbonate-gas purification followed by sponge iron and activated carbon traps should accomplish this goal.

In industrial practice, the BTX would be removed in the oil scrubber (fig. 1). A commercial plant (250,000,000 scfd) will produce about 25,000 gpd of BTX.

## TAR ANALYSIS

Tar analyses were made by our own and other laboratories, as shown in tables 4 and 5. Table 4 shows analyses of tars from various coals and the variety of products found in the tar.

Elemental analyses of tars are shown below, in percent:

| | Illinois No.6 | Illinois Char | Lignite |
|---|---|---|---|
| Carbon....... | 82.6 | 83.4 | 83.8 |
| Hydrogen... | 6.6 | 6.6 | 7.7 |
| Nitrogen..... | 1.1 | 1.1 | 1.0 |
| Sulfur......... | 2.8 | 2.6 | 1.1 |

These analyses are reported on a moisture- and ash-free basis. Oxygen can be obtained by difference from 100 percent. It is planned to burn the tars in the boiler because it would be too expensive to try to separate the chemical compounds for sale.

## SULFUR ANALYSIS

Sulfur analyses of the various coals tested and products are shown in table 5.

## SOLID ANALYSIS

The residue (char) from the gasifier in a commercial plant is to be burned along with the tars to raise steam for the process. The ultimate and proximate analyses of the chars are shown in table 6. The chars would contain a percentage of trace elements shown in ash analyses done by the Bureau (refs. 3,4). A problem may exist with $SO_x$ in stack gases when burning the char and tars from the gasification of high-sulfur coal.

## OTHER STREAMS

Since we operate both gasification and methanation laboratory-size equipment at PERC, we have analyzed these streams extensively. No serious contaminants have been detected in the water from the methanation reactors. The other process steps are not being investigated by PERC, but some assumptions can be made. It would be reasonable to assume that the contaminated condensate from the shift converter would be similar to, but more dilute, than the gasifier condensate shown in table 1. There should be no effluent from the hot carbonate unit if the feed gas contains the proper partial pressure of water. The Stretford unit should have a gas effluent low enough in sulfur for air pollution requirements for the 75-tpd plant at Bruceton.

## TRACE ANALYSIS

Other analyses that took special methods are shown in table 7. The HCN analysis is of special significance since it could effect the operation of the Stretford unit in the 75-tpd pilot plant. However, the low level

Table 7. Trace components in gas and tar

| | Gas (by volume) | | Tar (by weight) | |
|---|---|---|---|---|
| | HCN, ppb | Mercury, ppm | Mercury, ppm | Arsenic, ppm |
| Illinois char......... | 5 | - | - | - |
| Illinois No. 6 coal... | 20 | 0.00001 | 0.003 | 0.7 |
| Western Kentucky coal. | 11 | - | - | - |
| North Dakota lignite.. | 3 | - | - | - |
| Wyoming subbitum. coal. | 2 | - | - | - |

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*Reference to trade names is made to facilitate understanding and does not imply endorsement by the Bureau of Mines.

(ppb) indicates no serious problem. Mercury is present in the gas from the gasifier, but none was detected in the final product; that is, in the high-Btu gas. The mercury and arsenic in the tars, if burned, would probably end up in the stack gas.

## CONCLUSION

While we have obtained much data, we are continuing to analyze these effluent streams exhaustively. We hope to completely characterize these effluents and to have solutions available for handling them before the 75-tpd plant is operating. No serious problems are foreseen at this time.

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# CLEAN ENVIRONMENT WITH KOPPERS-TOTZEK PROCESS

**J. Frank Farnsworth,**
**D. Michael Mitsak,**
**and J. F. Kamody***

*Abstract*

*The Koppers-Totzek Process is the first commercially proven gasification process to be evaluated (in 1973) for the U.S. Environmental Protection Agency by Esso Research and Engineering Company.*

*This process converts any rank coal or heavy liquid hydrocarbon into a gas whose composition is essentially carbon dioxide, carbon monoxide, and hydrogen. Sulfur appears as hydrogen sulfide and is recovered in a Claus unit. Gas can be used as low-Btu (300) fuel; in reducing applications; as feedstock for chemical synthesis; or it can be upgraded to pipline quality. Oxygen and steam are introduced into the gasifier with the feedstock. This process produces only gas and slag; there are no liquid tars or hydrocarbons.*

## INTRODUCTION

As a result of recent shortages of clean gaseous and liquid fuels in the United States, numerous development programs have been initiated for the production of synthetic fuels from our indigenous natural resources. This activity has introduced into the American society a group of technical and nontechnical Americans, whom we identify as "Environmentalists." We find these concerned people among our friends, neighbors, and members of our families. They are a part of a group that is concerned with the protection of the environment from adverse affects of any future energy plants. In essence, their slogan could be, "Use the natural resources, but whatever residuals are returned to the earth must be of a purity equal to, or better than, the original material."

Gasification of solid or liquid fuels by the commercially proven Koppers-Totzek (K-T) process offers a pollution-free means of producing a 300 Btu per cubic foot synthesis gas which can be readily substituted for natural gas in industrial fuel applications. It can be catalytically upgraded to a heating value of 960 Btu per cubic foot to provide a gas comparable to, and interchangeable with, natural gas. The gas is an excellent base for the production of chemicals, such as ammonia and methanol. The K-T synthesis gas can also be converted to liquid hydrocarbons by the use of the well-known Fischer-Tropsch technology. Actually, this application gave birth in 1948 to the K-T process. At the time, the U.S. Bureau of Mines selected the K-T partial oxidation of pulverized coal in steam and oxygen process to supply the synthesis gas for coal-to-oil demonstration plant at Louisiana, Missouri. Koppers-Pittsburgh joined Koppers-Essen in developing the gasification technology from laboratory data into an operating unit designed for a gasification capacity of 36 tons of coal per day. Results at Louisiana were successful. Since 1952, Koppers-Essen has engineered and installed 33 gasifiers in 12 different locations throughout Europe, Africa, and Asia. Currently, 15 additional gasifiers at four locations are in various stages of construction. These plants, in general, use low-rank coals such as lignite or subbituminous, and the syn-gas is converted into ammonia. The rapid expansion of the natural gas and oil industries in the 1950's, with their abundant supplies of low cost gas and oil, relegated the K-T process to the shelf until the present energy shortage emerged.

To date, the Koppers-Totzek process is the only commercially proven gasification process to undergo a pollution evaluation study for the U.S. Environmental Protection Agency. Esso Research and Engineering Company, Linden, New Jersey, under contract to EPA, began the evaluation of the K-T process in early 1973. The completed report, titled "Evaluation of Pollution Control in Fossil Fuel Conversion Process, Gasification; Section 1: Koppers-Totzek Process, January 1974," has been issued and is available upon request through the Environmental Protection Agency, Research Triangle Park, North Carolina 27711. Of particular interest are the following excerpts taken from the report:

"This process can be used to make synthesis

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*J. Frank Farnsworth is senior staff engineer in coal and gas technology; D. Michael Mitsak is manager of the chemical project section; and J. F. Kamody is project engineer. They are with the Engineering and Construction Division of the Koppers Company, Inc., Pittsburgh, Pennsylvania.

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> gas, reducing gas, or fuel gas, and was studied first for several reasons: (1) more complete information is available than on some other processes; this specific design does not include proprietary clean-up processes; and there are a number of commercial plants in operation; (2) it is a simple and relatively clean process in that it does not produce tar, oil, or phenols, (Minor amounts of cyanide, ammonia, etc., are produced); (3) the process developer was cooperative in supplying requested information."
>
> "Low-Btu gas, such as that from the Koppers-Totzek gasifier, would be expected to give lower $NO_x$ due to lower flame temperature." (This refers to comparison with natural gas and coal firing.)

Although EPA has reviewed and approved the report for publication, the approval is qualified as follows: "Approval does not signify that the contents necessarily reflect the views and policies of the agency, nor does the mention of trade names or commercial products constitute endorsement or recommendation for use." In our opinion, this report must be considered a base for the position that regulatory agencies may take, since it indicates what is attainable with present technology.

## PROCESS DESCRIPTION AND PROCESS FEATURES

The total process for producing clean, desulfurized utility gas or synthesis gas is shown schematically (figure 1). Depending on rank, the coal is dried to between 2 and 8 percent moisture and is pulverized to about 70 percent through 200 mesh (figure 2). The coal is conveyed with nitrogen from storage to the gasifier service bins. Controls regulate the intermittent feeding of coal from the service bins to the feed bins, which are connected to variable-speed coal screw feeders (figure 3). The pulverized coal is continuously discharged into a mixing nozzle where it is entrained in oxygen and low pressure steam. Moderate temperature and high burner velocity prevent the reaction of the coal and the oxygen until entry into the gasification zone.

The oxygen, steam, and coal react in the gasifier (figure 4) at a slight positive pressure and at 3,300°F. The carbon and volatile matter of the coal are gasified, and the coal ash is converted into molten slag. About 50 to 70 percent of this slag drops into a water quench tank and is carried from the tank to the plant disposal system as a granular solid, and the remainder is entrained in the gas exiting the gasifier. Low pressure steam for the gasifier reaction is produced in the gasifier jacket from the heat passing through the refractory lining.

Gas leaving the gasifier is direct-water quenched when it is necessary to solidify entrained slag droplets and then passed through a waste heat boiler where high pressure steam up to 1,500 psig is produced. After leaving the waste heat boiler, the gas is cleaned and cooled in a venturi scrubbing system in which the entrained solids are reduced to 0.002 to 0.003 grains per SCF, and the temperature lowered from 350°F to about 95°F. Electrostatic precipitators are used to reduce particulates to 0.0001 grains per SCF, if the K-T gas is to be compressed to high pressures for chemical synthesis or for the production of high-Btu gas. Several gasifiers can share common cleaning and cooling equipment.

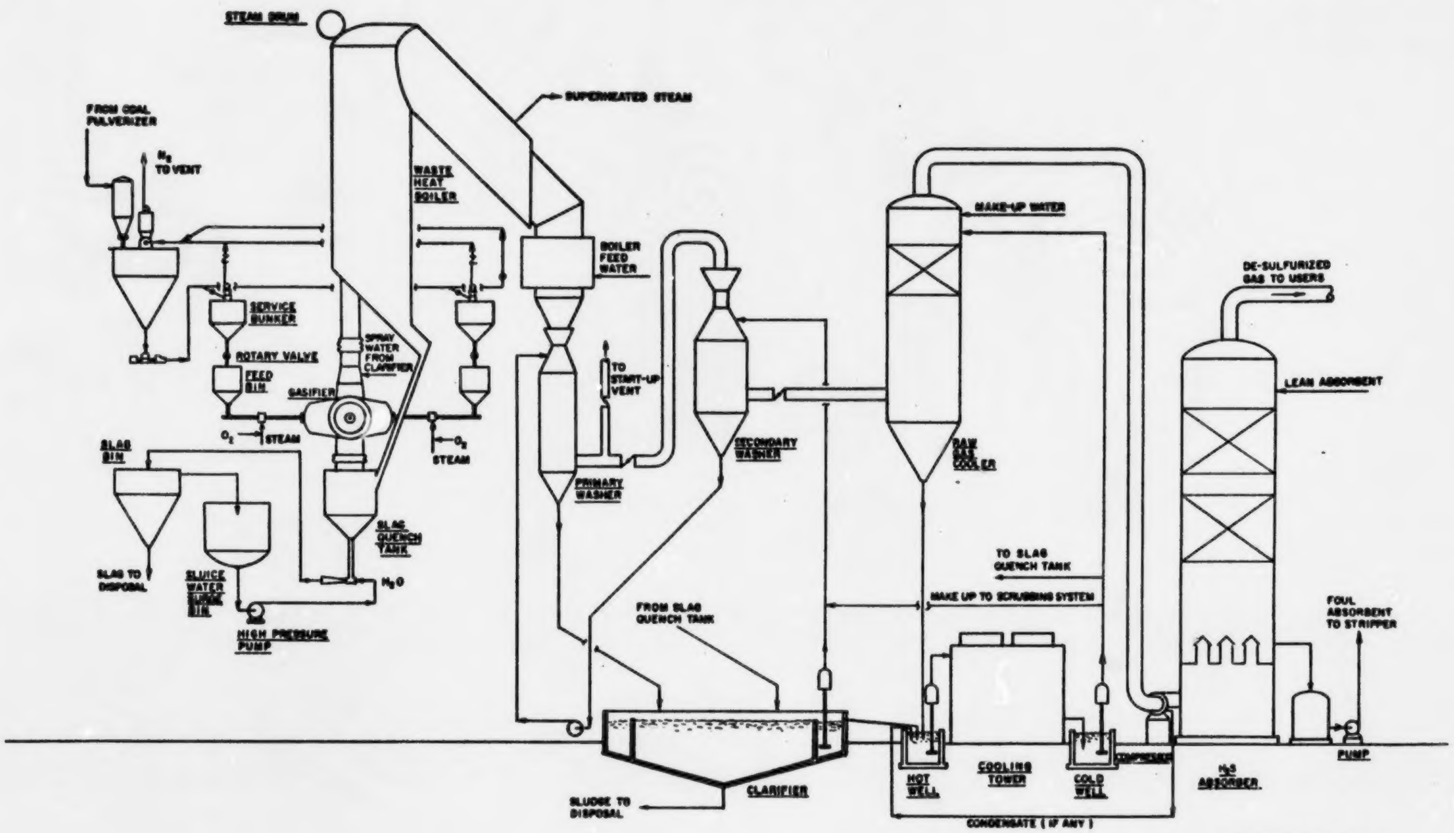
Particulate-laden water from the gas cleaning and cooling system is piped to a clarifier. Sludge from the clarifier is pumped either to a filter or to the plant disposal area. Clarified water is recirculated through the venturi scrubbers and the excess overflows into the cooling tower system at the gas cooler. Evaporation, windage, and blowdown water losses at the cooling tower, plus moisture in clarifier sludge and in slag, necessitate the addition of a small quantity of makeup water to this system. If water is at a premium, air cooling may be used for certain applications, and the cooling tower can be reduced in size to provide only the final trim in water temperature.

The cool, clean gas leaving the gas-cleaning system contains sulfur compounds which must be removed to meet gas specifications. The type of system chosen depends upon the end uses and pressure of the product gas. For low pressures (up to 150 psig) and low-Btu gas application, there are the chemical reaction processes such as amine and carbonate systems. At higher pressures, the physical absorption processes such as Rectisol, Purisol, and Selexol are used. The choice of the process is also dependent upon the desired purity of the product gas and the desired selectivity with respect to the concentrations of carbon dioxide and sulfides.

Tables 1 & 2 show typical gasification data for western and eastern coals and petroleum coke feedstock.

The K-T process offers advantages not available in existing nonsuspension systems. In the gasification of

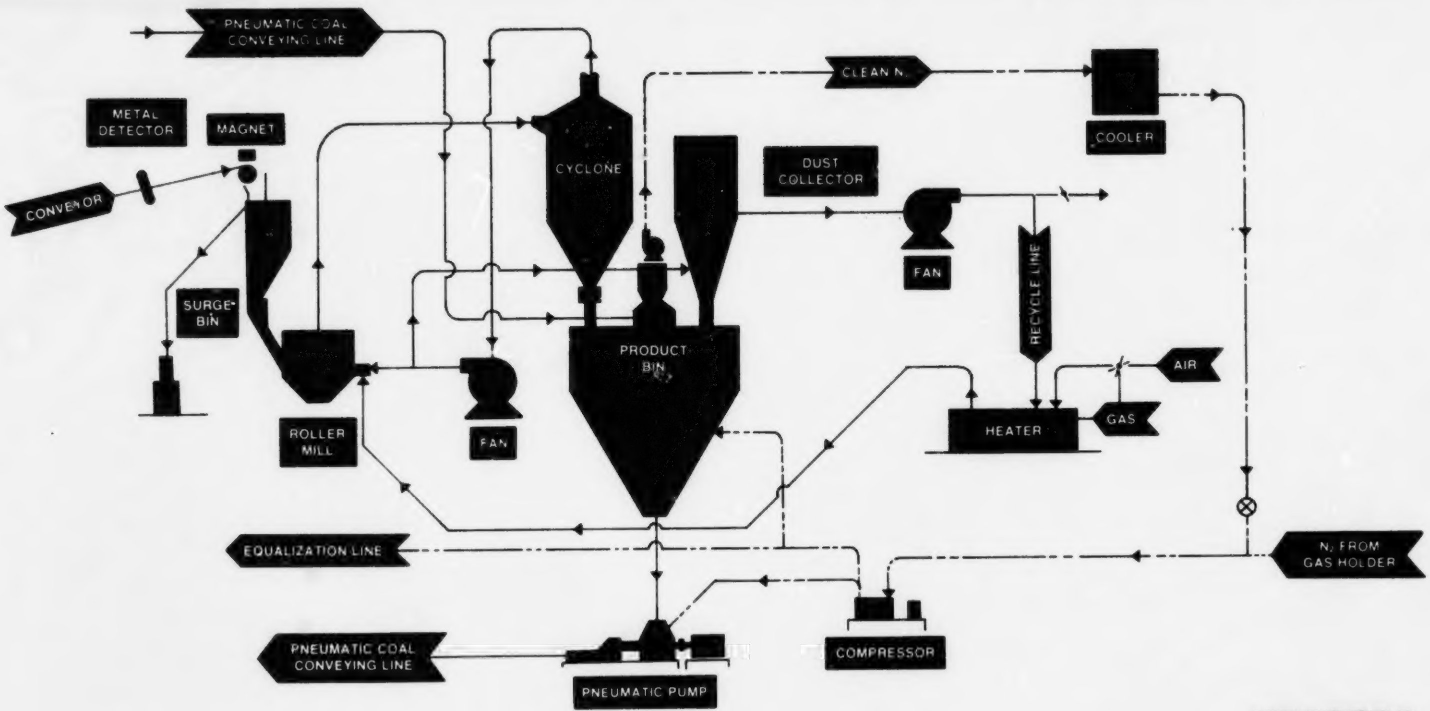
# K-T Gasification Process


STEAM DRUM
FROM COAL PULVERIZER
$N_2$ TO VENT
SUPERHEATED STEAM
WASTE HEAT BOILER
SERVICE BUNKER
ROTARY VALVE
FEED BIN
GASIFIER
SPRAY WATER FROM CLARIFIER
$O_2$
STEAM
$O_2$
STEAM
SLAG BIN
SLAG TO DISPOSAL
SLUICE WATER SURGE BIN
HIGH PRESSURE PUMP
SLAG QUENCH TANK
$H_2O$
BOILER FEED WATER
TO START-UP VENT
PRIMARY WASHER
SECONDARY WASHER
MAKE-UP WATER
RAW GAS COOLER
DE-SULFURIZED GAS TO USERS
LEAN ABSORBENT
TO SLAG QUENCH TANK
MAKE UP TO SCRUBBING SYSTEM
FROM SLAG QUENCH TANK
FOUL ABSORBENT TO STRIPPER
SLUDGE TO DISPOSAL
CLARIFIER
HOT WELL
COOLING TOWER
COLD WELL
COMPRESSOR
$H_2S$ ABSORBER
PUMP
CONDENSATE ( IF ANY )

**KOPPERS**

FIGURE 1

# COAL PREPARATION


PNEUMATIC COAL CONVEYING LINE
METAL DETECTOR
MAGNET
CONVEYOR
SURGE BIN
ROLLER MILL
FAN
CYCLONE
PRODUCT BIN
CLEAN $N_2$
DUST COLLECTOR
FAN
COOLER
RECYCLE LINE
AIR
GAS
HEATER
EQUALIZATION LINE
$N_2$ FROM GAS HOLDER
PNEUMATIC COAL CONVEYING LINE
PNEUMATIC PUMP
COMPRESSOR

KOPPERS

FIGURE 2

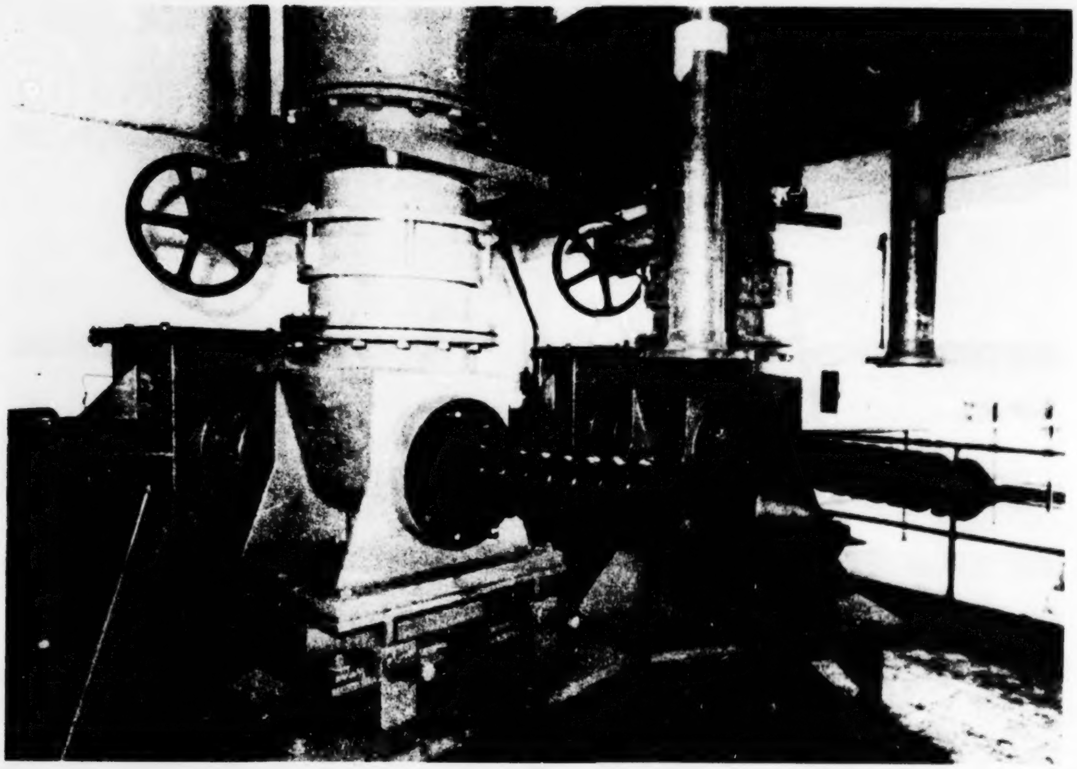


FIGURE 3. COAL SCREW FEEDER

coal, the entire mine output is usable. Coal size is not a limiting factor, and coking coals can be handled without pretreatment. Other features include:

*Versatility*. The process is capable of continuous operation for the gasification of a variety of feedstocks, including all ranks of solid fuels, liquid hydrocarbons, and pumpable slurries of carbonaceous materials in liquid hydrocarbons. Feedstocks of high ash and sulfur contents, not environmentally acceptable in present energy plants, can be readily used in the K-T process.

*Flexibility*. The change-over from solid to liquid fuels involves only a change of the burner heads. Multiple feed burners and variable speed screw feeders permit turndown to 70 percent. The process is capable of instantaneous shutdown; from hot standby, full gas production is resumed in less than 30 minutes.

*Simplicity of Construction and Ease of Operation*. The only moving parts at the gasifier are screw feeders for solids, or a pump for liquid feedstocks. Control of the gasifiers is achieved primarily by maintaining $CO_2$ concentration in the clean gas at a reasonably constant and predetermined value. Slag fluidity may be visually monitored. Gasifiers display good dynamic response.

*Low Maintenance*. Simplicity and a minimum of moving parts result in little maintenance between scheduled annual shutdowns for boiler inspection. The gasifier is lined with a monolithic castable refractory which lasts as long as 5 years with a minimum of patching.

*High Capacity*. K-T units are designed for coal feed rates of up to 850 tons per day, or for a production of about 45 million standard cubic feet per day of 300-Btu gas.

*Safe and Efficient*. The K-T process has over 20 years of safe operation. The onstream time or availability is better than 95 percent. Heating value of the product gas is equivalent to 70-77 percent of the calorific value of the feedstock. Additional energy recovered as steam from the available sensible heat

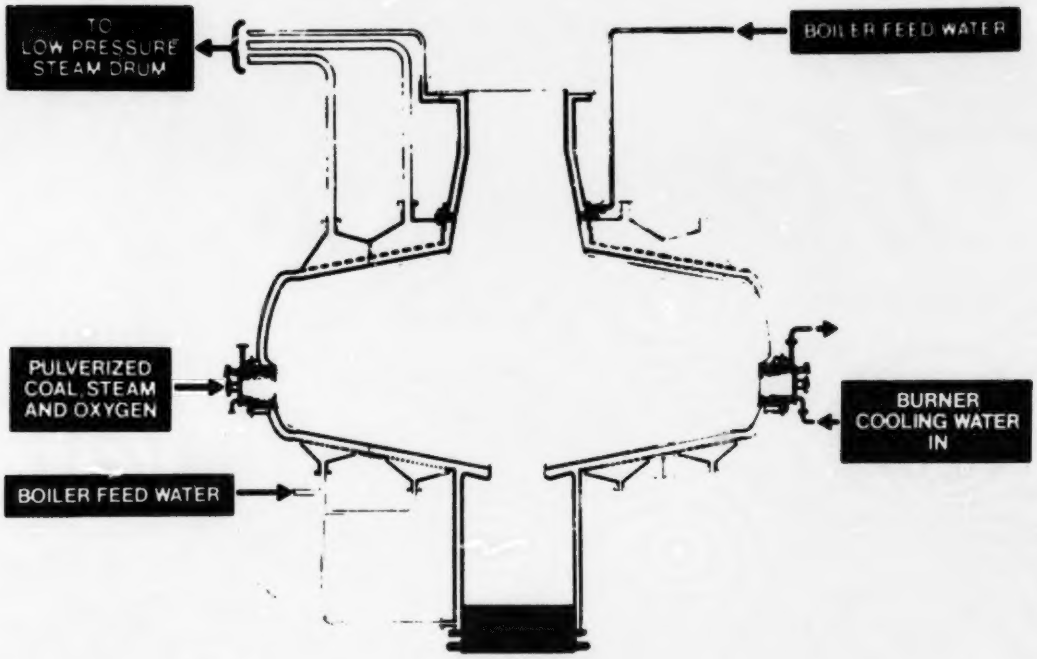


FIGURE 4

amounts to about 15 percent of the calorific value of the coal.

## ENVIRONMENTAL CONTROLS

Control of the environment in a fuel conversion facility depends, to a great degree, upon the controlability and simplicity of process operations. The 25-year K-T record of commercial operation shows a consistency in continuous production at 95 percent of the onstream design figure. This reliance requires controlability. As noted in the aforementioned pollution evaluation report, "the process is simple and clean." This is possible since the products from the Koppers-Totzek gasifier are slag and a synthesis gas composed primarily of carbon dioxide, carbon monoxide, and hydrogen. The sulfur contained in the feed material is converted during gasification to hydrogen sulfide and carbonyl sulfide. The sulfur compounds can be removed from the gas to regulatory environmental limits with commercially proven processes, and converted to elemental sulfur suitable for sale to the chemical industry. The particulate matter in the gas is removed by water scrubbing the gas in two stages of venturi scrubbers. The slag as produced is granulated and, since it has passed through the molten state in the gasifier, contains little or no dust or leachable material. The slag will be suitable for road aggregate, landfill, or for use in cinder blocks. The trace amounts of ammonia, cyanide, etc., in the raw gas are removed during gas cleaning and disposed of by combustion in the Claus sulfur unit. The clean gas can be burned in conventional power-generating equipment, and the combustion gases are acceptably low in $NO_x$ and other atmospheric pollutants.

### *Coal Storage and Handling*

Gasification plants require an emergency supply of coal. To minimize air pollution due to coal dust, this coal supply will be placed in "dead storage." The term "dead storage" means that this quantity of coal is held in a compacted and sealed pile not susceptible to dusting during wind activity. The coal storage pile is prepared by layering coal in 12-inch depths and by compacting each layer to a bulk density of about 70 pounds per cubic foot. The height of the pile is set at about 25 feet, and the length and width are fixed by the tonnage to be stored. In order to monitor

## TABLE I

## K-T GASIFIER DATA FOR VARIOUS FUELS

| TYPE OF FUEL | WESTERN COAL | EASTERN COAL | GREEN PETROLEUM COKE |
|---|---|---|---|
| **GASIFIER FEED** | | | |
| Dry Feed to Gasifier | | | |
| Analysis, Wt% | | | |
| C | 72.7 | 69.9 | 88.0 |
| H | 5.3 | 4.9 | 4.5 |
| N | 1.1 | 1.3 | 1.4 |
| S | 1.0 | 1.1 | 4.3 |
| O | 9.0 | 7.1 | 1.0 |
| Ash | 8.9 | 13.7 | 0.2 |
| Moisture | 2.0 | 2.0 | 0.6 |
| | 100.0 | 100.0 | 100.0 |
| Gross Heating Value of Dry Feed, Btu/Lb | 13,135 | 12,640 | 15,690 |
| Oxygen, Tons/Ton Dried Feed @ 98% Purity | 0.878 | 0.849 | 0.950 |
| Process Steam, Lb/Ton Dried Feed | 814 | 810 | 1182 |
| **GASIFIER PRODUCTS** | | | |
| Jacket Steam, Lb/Ton Dried Feed | 600 | 554 | 629 |
| High Pressure Steam, Lb/Ton Dried Feed @ 900°F/900 psig | 2760 | 2675 | 3598 |
| Raw Gas Analysis, Vol%, Dry | | | |
| CO | 52.55 | 52.51 | 52.22 |
| $CO_2$ | 10.00 | 10.00 | 10.00 |
| $H_2$ | 36.09 | 35.96 | 35.40 |
| $N_2$ + Argon | 1.00 | 1.15 | 1.10 |
| $H_2S$ | 0.34 | 0.36 | 1.20 |
| COS | 0.02 | 0.02 | 0.08 |
| | 100.00 | 100.00 | 100.00 |
| Dry Gas Make - SCF/Ton Dried Feed | 69,690 | 66,970 | 77,500 |
| Gas Gross Heating Value, Btu/SCF, Dry | 287 | 286 | 283 |
| % Heating Value of Gas/Heating Value Feed (Gross Basis) | 76.1 | 75.8 | 69.9 |

spontaneous combustion, thermocouples are inserted throughout the pile. The outer surface of the pile is sprayed with an organic polymer crusting agent to prevent dusting or rain erosion. Crusting also prevents rainwater penetration of coal particles; thus, water runoff will have little or no contaminants such as those found in mine waters. In addition, the coal pile will be located on a waterproof base to prevent water seepage into the ground. Thus, all runoff water will be contained and used in the process. Under this arrangement, the daily in-and-out requirements of coal transfers will be performed in totally enclosed equipment, and coal from "dead storage" will be taken only in an emergency when the normal supply of coal is interrupted.

In normal practice, coal will be delivered to the plant by rail (figure 5). To economically deliver the large daily tonnages of coal required by gasification

## TABLE 2

## K-T GASIFIER HEAT BALANCE

Western Coal (2% Moisture)

Gas Exit Temperature = 2730°F
Reference State = Liquid Water

| HEAT INPUT - ABOVE 60°F | Btu/Lb of Coal Feed | % |
|---|---|---|
| Calorific Value of Coal | 13,135 | 96.43 |
| Sensible Heat in Coal @ 160°F | 29 | 0.21 |
| Sensible Heat in Oxygen @ 230°F | 27 | 0.20 |
| Total Heat in Steam @ 250°F | 430 | 3.16 |
| TOTAL | 13,621 | 100.00 |

| HEAT OUTPUT - ABOVE 60°F | Btu/Lb of Coal Feed | % |
|---|---|---|
| Heat in Molten Slag and Ash | 67 | 0.49 |
| Sensible Heat in Unburned Carbon | 36 | 0.26 |
| Calorific Value of Unburned Carbon | 527 | 3.87 |
| Calorific Value of Gas | 9,996 | 73.39 |
| Sensible Heat Dry Gas | 1,994 | 14.64 |
| Total Heat Water Vapor in Gas | 712 | 5.23 |
| Total Heat - Gasifier Jacket Steam | 275 | 2.02 |
| Gasifier Heat Loss | 14 | 0.10 |
| TOTAL | 13,621 | 100.00 |

plants, unit trains hauling 100-125 net ton capacity cars, will be used. Loading facilities at the mine and unloading facilities at the plant will be designed to accommodate a train delivering coal on a regular schedule, thus eliminating the need of large active storage facilities with their attendent pollution problems. Tracks at the mine and at the plant will be designed with loops to reverse the train direction at each end, without switching.

Mine facilities will include: a loop spur capable of holding an entire train while it is being loaded; coal handling, crushing, and storage systems for delivering a 2" x 0" product; an overtrack tipple designed to allow flood-loading of the cars without stopping the train; and a plant-leg chute to transfer coal flow from one car to the next with a minimum of spillage between cars. Special flat-bottomed cars with "flip-top" covers can be provided to exclude the elements during transit, and to prevent windblown loss of coal dust and the resultant air pollution.

At the plant site, a track loop capable of holding an entire train while it is being unloaded will be provided. The coal will be dumped by a rotary car dumper into a double compartment track hopper which is completely housed, and an automatic car positioner will be provided that will be capable of moving the entire loaded train, including the locomotive, as a unit. The car dumper, car positioner, and crusher will be housed in a structure that will have dust-collecting equipment to control particulate emission to the atmosphere at acceptable limits. Sound absorbent panels will be used where practical and effective to control the noise level. Cars with lids will automatically open for dumping and will automatically close before leaving the structure. The cars will be emptied and shifted at about 25 to 28 cars per hour. At this rate, a complete trainload of 12,000 tons of coal can be processed in less than 5 hours.

Under normal operating conditions, coal will be conveyed from the track hopper directly to two rows of concrete vertical, cylindrical, cone-bottomed, closed, storage bins, and each row will contain about a day's supply of coal. Coal will be delivered to each row of bins by covered conveyors. A house will be provided over the top of each row of bins. Each bin,

## COAL HANDLING, STORAGE & PREPARATION

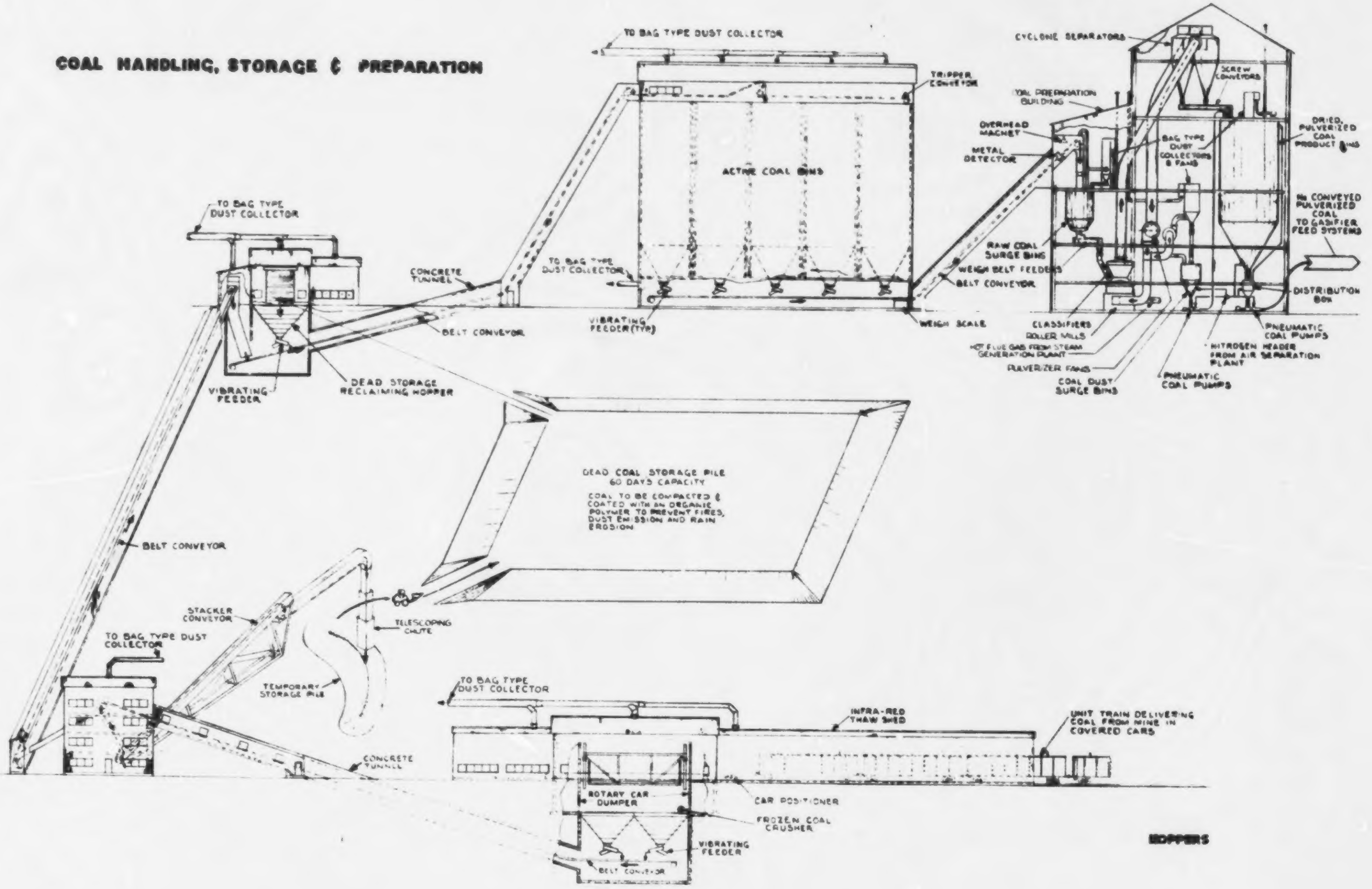

TO BAG TYPE DUST COLLECTOR
TRIPPER CONVEYOR
ACTIVE COAL BINS
TO BAG TYPE DUST COLLECTOR
VIBRATING FEEDER (TYP)
WEIGH SCALE
TO BAG TYPE DUST COLLECTOR
CONCRETE TUNNEL
BELT CONVEYOR
VIBRATING FEEDER
DEAD STORAGE RECLAIMING HOPPER
CYCLONE SEPARATORS
SCREW CONVEYORS
COAL PREPARATION BUILDING
OVERHEAD MAGNET
METAL DETECTOR
BAG TYPE DUST COLLECTORS & FANS
DRIED, PULVERIZED COAL PRODUCT BINS
N₂ CONVEYED PULVERIZED COAL TO GASIFIER FEED SYSTEMS
RAW COAL SURGE BINS
WEIGH BELT FEEDERS
BELT CONVEYOR
DISTRIBUTION BOX
CLASSIFIERS
ROLLER MILLS
HOT FLUE GAS FROM STEAM GENERATION PLANT
PULVERIZER FANS
COAL DUST SURGE BINS
PNEUMATIC COAL PUMPS
NITROGEN HEADER FROM AIR SEPARATION PLANT
PNEUMATIC COAL PUMPS
DEAD COAL STORAGE PILE 60 DAYS CAPACITY
COAL TO BE COMPACTED & COATED WITH AN ORGANIC POLYMER TO PREVENT FIRES, DUST EMISSION AND RAIN EROSION
BELT CONVEYOR
STACKER CONVEYOR
TELESCOPING CHUTE
TO BAG TYPE DUST COLLECTOR
TEMPORARY STORAGE PILE
CONCRETE TUNNEL
TO BAG TYPE DUST COLLECTOR
INFRA-RED THAW SHED
UNIT TRAIN DELIVERING COAL FROM MINE IN COVERED CARS
ROTARY CAR DUMPER
CAR POSITIONER
FROZEN COAL CRUSHER
VIBRATING FEEDER
BELT CONVEYOR
KOPPERS

FIGURE 5

and the house itself, will be connected to a dust collecting system to control dust emission and to negate the possibility of fires or explosions. In addition, a vacuum-cleaning system will be provided in these houses.

All conveyors will be completely enclosed in galleries, and all conveyor junction towers will have junction houses to completely enclose the coal transfer points. Each junction house will have a dust collection system.

### *Coal Preparation*

The coal preparation facilities start with surge bins which are fed by a totally enclosed belt-conveying system originating at the concrete coal storage day bins. Coal from a surge bin is delivered to a pulverizing, classifying, and drying system in which the coal is pulverized to 70 percent minus 200 mesh and dried to 2 percent moisture. Flue gas at 800°F produced by the combustion of clean process gas is used as the drying medium. The flue gas intimately contacts the wet coal in a fluidized or entrained operation and drives off the moisture. At these conditions, the coal temperature does not exceed 180°F; as a result, there are no chemical reactions or devolatilization of the coal. Thus, the flue gas and water vapor will not be contaminated and can be discharged to the atmosphere as pollution free after separation from the dried and pulverized coal. Particulate matter in the discharged gases will be controlled to acceptable limits by the use of cyclones, bag filters, or precipitators. Condensation of the water vapor from the stack could be controlled by the introduction of combustion gases from other parts of the plant, or by the use of tall stacks to dissipate the vapors.

The conveyors and chutes feeding the surge bins will be enclosed, along with the weigh belt feeders and chutes feeding the roller mills. These housings will be connected to a dust collecting system. The product bins will be connected to a common bag-type dust collector; individual dust collectors, vented back to the product bins via the pneumatic conveying system lines, will be provided for each service bunker. The feed systems delivering dust from the pulverized bag filters to the surge bins will be enclosed and vented back to the dust collectors. The entire coal preparation facility, starting with the pulverized mill surge bins and including the product bins, will be housed in a building constructed of sound-absorbent panels where practical, to reduce noise level. Where possible, operating equipment will be designed with noise suppression features. The building ventilating system will discharge through an additional bag filter in order to control dust leaving the building. Also, a vacuum cleaning system will be provided for the facility to further insure clean working conditions for the operators.

Since the entire coal preparation facility is enclosed in an essentially dust-tight building, the only exit for dust to the atmosphere will be the building ventilating dust collector. Particulates entering this collector will be minimal because of the equipment dust collectors within the building. Thus, the particulate emission to the atmosphere from coal preparation will not exceed the designated dust loading.

Water requirements in the coal preparation area will be limited to those used for cooling of bearings; therefore, this facility does not add any polluting products to the plant water systems.

The plant will be designed to reduce the noise level at the plant boundary to 50 dB(A).

### *Gasification, Gas Cooling, and Cleaning*

As mentioned earlier, the K-T process, due to the high temperature (3,330-3,500°F) operations, produces gas and slag. Because there are no condensible hydrocarbons, possible pollution sources are limited to the gas, slag, process waters, and/or gas condensates. Environmentally, by today's EPA standards, objectionable gaseous matter from any plant includes particulate matter, cyanides, sulfur compounds, and oxides of nitrogen. Water-contaminating substances include oil, and such serious taste-offending compounds as phenols and pyridines. The K-T process does not produce phenols, pyridines, or organics; however, ammonia and cyanide are produced in amounts well under one volume percent. This situation provides for minimal treatment to meet EPA discharge standards.

The following gas analyses (table 3) at the various processing stages indicate the possible contaminants and their concentrations.

The reduction in the particulate matter from 12 grains/SCF to 0.002 grains/SCF in gas occurs in the primary and secondary venturi scrubbers where the gas is intimately contacted with recirculated water. In the process the water soluble ammonia, sulfur dioxide, hydrogen cyanide, and some hydrogen sulfide are absorbed. A bleed stream from the recirculated water system is continuously discharged to a stripper, where the gaseous components are stripped by vapor rising from a steam reboiler. Gaseous $NH_3$,$SO_2$, and HCN flow to the Claus unit and are combusted while the stripped water is cooled

TABLE 3

| | VOLUME PERCENT | | |
|---|---|---|---|
| Component | Gasifier Outlet | To Compression & Acid Gas Removal | Product Gas |
| CO | 37.36 | 49.50 | 53.16 |
| $CO_2$ | 7.13 | 9.42 | 9.44 |
| $CH_4$ | 0.08 | 0.11 | 0.12 |
| $H_2$ | 25.17 | 33.35 | 36.51 |
| $N_2$ | 0.30 | 0.40 | 0.44 |
| $H_2S$ | 0.23 | 0.30 | 3 ppmv |
| COS | 178 ppmv | 235 ppmv | 1.5 ppmv |
| HCN | 288 ppmv | 300 ppmv | 1 ppmv |
| $NH_3$ | 0.17 | 0.22 | 1 ppmv |
| $H_2O$ | 29.19 | 6.20 | 160 ppmv |
| Ar | 0.32 | 0.42 | 0.46 |
| $SO_2$ | 22 ppmv | 15 ppmv | 0.5 ppmv |
| No | 7 ppmv | 7 ppmv | 3 ppmv |
| Particulates (grs/SCF) | 11.57 | <0.002 | <0.002 |

and returned to the stripper-water-circulating system. Excess stripped water is bled to the boiler feedwater system where it is treated and used in the generation of steam at the gasifiers.

The particulate matter (entrained slag) remaining in the water that passes from the venturi scrubbers is removed in a clarifier. If water is not at a premium, the particulate in slurry form can be pumped to a disposal area outside the plant. When water is scarce, the slurry would be filtered, the water returned to scrubbers, and the filter cake placed in loadout bins for truck or railroad car disposal. Since particulate matter is slagged material, there should be no leachable contaminants, and disposal should pose no environmental problems. The water recirculating system is pollution clear with the side stream treatment arrangement. The cooling of the recirculating water can be achieved on a direct basis in a cooling tower, if this is acceptable environmentally, or on an indirect basis with air or water exchangers.

The following water analyses (table 4) of the various gas cooling and cleaning steps at the Kutahya, Turkey, plant were reported by the Kutahya personnel in late 1972. The data is offered to indicate what order of magnitude that concentrations are possible, and identifies possible contaminants. It is to be noted that the most objectionable contaminants, phenols, tars, oils, and pyridines are missing.

As shown in table 4, solubility of hydrogen sulfide in water is slight; thus, the gas leaving the venturi scrubbers contains most of the hydrogen sulfide and carbonyl sulfide. There are various acid gas removal processes that have the capability of reducing the sulfur content in the gas to 5 ppmv. The processes are based on absorption in solution and subsequent stripping of the acid gases of $H_2S$ and $CO_2$ from the solution. The physical absorption processes, which operate at pressures of 300-400 psig, exhibit the greatest selectivity for hydrogen sulfide and carbon dioxide removal. Since no chemical reactions occur, these processes do not form stable compounds such as thiosulfates and thiocyanates. Some chemical reaction processes, such as carbonate and amine, which form the aforementioned stable compounds, can be used but will require periodic dumping of the solution in order to maintain removal efficiency. Dumped solution will require treatment to meet permissible discharge limitations. The choice of process is dependent upon economics, environmental control, purity of product gas, and desired acid gas selectivity. A plant can be designed to reduce sulfur in product gas to 5 ppmv, to control the $H_2S$ level in carbon dioxide in 10 ppmv, and to reduce the liquid effluent to zero pollutants.

The acid gas stream, containing a minimum of 14 volume percent $H_2S$, is catalytically converted to elemental molten sulfur in a Claus unit. The tail gases exiting the Claus unit contain $SO_2$ and can be treated to catalytically reduce the $SO_2$ to $H_2S$. Scrubbing with an amine solution absorbs the $H_2S$ and subsequent stripping yields an $H_2S$ stream which is recycled to the Claus unit. This combination results in overall sulfur recovery of 99+percent.

At this stage, the clean gas is available as fuel or as feedstock for further processing to chemicals and to pipeline (high-Btu) gas. As regards chemical usage, the gas itself does not add to environmental considerations. On the other hand, if the gas is used as a source of fuel for inplant use or in combined cycle power generation, then consideration must be given to the combustion characteristics and $NO_x$ formation.

Gases containing carbon monoxide and hydrogen, such as coke oven gas, producer gas, water gas, and blast furnace gas, have been used as industrial fuels for many years. American manufacturers of modern steam boilers and gas turbines foresee no major problems in adapting furnaces and combustors to utilize the K-T utility gas.

When comparing the calorific value of the K-T utility gas with that of natural gas (300 versus 1,000 Btu), it is often misconceived by some people that the combustion characteristics of the K-T gas are

## TABLE 4

| Sample Location | | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
|---|---|---|---|---|---|---|---|---|
| pH Value | | 8.8 | 8.8 | 8.9 | 7.5 | 8.8 | 8.9 | 8.9 |
| Conductivity | S | 7.6<br>$10^{-4}$ | 1.8<br>$10^{-3}$ | 2.0<br>$10^{-3}$ | 9.7<br>$10^{-4}$ | 1.8<br>$10^{-3}$ | 1.8<br>$10^{-3}$ | 1.7<br>$10^{-3}$ |
| p Value | mval/l | 0.4 | 2.4 | 4.0 | 0.8 | 1.6 | 2.4 | 2.2 |
| m Value | mval/l | 5.8 | 13.2 | 14.0 | 6.4 | 12.8 | 14.8 | 14.0 |
| Total Hardness | ° dH | 20.8 | 33.5 | 36.8 | 22.8 | 34.0 | 34.8 | 35.2 |
| CaO | mg/l | 78 | 101 | 78 | 85 | 135 | 179 | 179 |
| MgO | mg/l | 97 | 161 | 194 | 102 | 145 | 113 | 129 |
| Na | mg/l | 17.5 | 17.5 | 17.5 | 17.5 | 17.5 | 17.5 | 17.5 |
| K | mg/l | 5.6 | 8.8 | 10.0 | 6.8 | 8.0 | 8.0 | 7.9 |
| Zn | mg/l | 0.01 | 0.03 | 0.02 | 0.03 | 0.02 | 0.02 | 0.02 |
| Fe | mg/l | 0.05 | 0.22 | 1.95 | 0.26 | 0.20 | 0.64 | 0.24 |
| $NH_4$ | mg/l | 0.32 | 157 | 184 | 25 | 137 | 122 | 72 |
| $NO_2$ | mg/l | 0.02 | 0.13 | 4.47 | 5.34 | 0.24 | 4.37 | 23.7 |
| $NO_3$ | mg/l | 58.2 | 3.32 | 13.7 | 34.0 | 24.7 | 22.9 | 42.0 |
| $PO_4$ Total | mg/l | 1.89 | 0.81 | 1.21 | 1.69 | 0.81 | 2.70 | 2.41 |
| Cl | mg/l | 18 | 85 | 96 | 53 | 57 | 46 | 36 |
| $SO_4$ | mg/l | 42 | 216 | 155 | 147 | 255 | 109 | 153 |
| CN | mg/l | 0.26 | 0.52 | 12.5 | 7.0 | 1.4 | 14.0 | 0.7 |
| $H_2S$ | mg/l | Not Detected | | | | | | |
| $KMnO_4$ Consumed | mg/l | 8 | 9 | 400 | 14 | 11 | 145 | |
| COD | $mgO_2/l$ | 14 | 18 | 128 | 18 | 16 | 63 | 60 |
| $SiO_2$ | mg/l | 14.8 | 16.0 | 14.8 | 30.6 | 19.8 | 42.6 | 30.6 |
| Suspended Solids | mg/l | 14 | 4612 | 5084 | 278 | 3072 | 50 | 58 |
| Hot Residue, 800°C | mg/l | 4 | 3918 | 4356 | 134 | 2690 | 46 | 42 |
| Stripped Residue | mg/l | 568 | 812 | 940 | 606 | 706 | 724 | 828 |
| Hot Residue, 800°C | mg/l | 268 | 550 | 588 | 366 | 526 | 512 | 534 |
| Cu | mg/l | 0.01 | 0.01 | 0.01 | 0.06 | 0.01 | 0.06 | 0.27 |

1) Cooling water to gasifier seal pot.

2) Water from the gasifier seal pot.

3) Wash water after washer-cooler.

4) Wash water after Theisen washer.

5) Water into clarifier.

6) Water out of clarifier.

7) Water out of cooling tower.

inferior to those of natural gas. Actually, the K-T gas displays excellent combustion characteristics resulting in boiler or gas turbine performances equivalent to, and in some ways better than, those for natural gas or for other utility gases produced from alternate coal gasification processes.

Table 5 compares the theoretical combustion characteristics of the K-T gas with a typical natural gas and with gas from an airblown fixed bed gasifier, as well as with a typical second generation coal gasification process (Bi-Gas). In each instance, the gases are used for firing a utility boiler. Comparison of the fuels is based on a total of one million Btu of net calorific heat input (measured at 60°F, 30 in. Hg). The net (or lower) heating value of the gas has been selected for comparison, since the latent heat of water vapor in the combustion gas is not recovered in combustion equipment. Air, initially at a 40°F wet bulb temperature, is compressed and enters the combustor at 600°F. For comparative purposes, all fuels are based on a delivery temperature of 77°F to the combustor, even though waste heat from a coal gasification process can often be used to preheat these fuels.

As can be seen in table 5, combustion of K-T gas requires only about 75 percent of the quantity of air required for theoretical combustion of the natural gas. The weight of the products of combustion from the K-T gas is about 95 percent of that which results from natural gas combustion, or combustion of gas from the BiGas Process. Thus, if these gases are fired to steam boilers at a given calorific heat input with

## Table 5. Combustion Characteristics of Various Fuels in Gas Turbine Application

| | High Btu Natural Gas | Bi-Gas | Air-Blown Fixed Bed | K-T Utility Gas |
|---|---|---|---|---|
| Composition, Vol% | | | | |
| $CO_2$ | 0.50 | 19.40 | 14.00 | 9.25 |
| CO | -- | 30.10 | 16.00 | 53.00 |
| $H_2$ | -- | 40.15 | 25.00 | 36.40 |
| $N_2$ + Argon | 0.50 | 0.68 | 39.70 | 1.05 |
| $CH_4$ | 83.00 | 9.37 | 5.00 | -- |
| $C_2H_6$ | 16.00 | -- | -- | -- |
| $H_2O$ | -- | 0.30 | 0.30 | 0.30 |
| | 100.00 | 100.00 | 100.00 | 100.00 |
| Total Lb-Moles of Dry Gas | 2.59 | 9.01 | 15.90 | 9.74 |
| Lower Heating Value, Btu/SCF, Dry | 1020 | 293 | 166 | 271 |
| Higher Heating Value, Btu/SCF, Dry | 1127 | 322 | 183 | 289 |
| DATA ON COMBUSTION WITH THEORETICAL AIR | | | | |
| Lb-Moles Air Required | 27.37 | 23.13 | 23.12 | 20.77 |
| Lb-Moles Theoretical Combustion Gas | 30.16 | 28.98 | 35.76 | 26.16 |
| Lb. of Theoretical Combustion Gas | 837 | 843 | 1035 | 794 |
| Adiabatic Maximum Flame Temp. °F | 4075 | 4000 | 3400 | 4280 |
| Composition of Complete Combustion Gas, Vol% | | | | |
| $CO_2$ | 9.90 | 18.30 | 15.56 | 23.19 |
| $N_2$ + Argon | 71.65 | 63.20 | 68.67 | 63.05 |
| $H_2O$ | 18.45 | 18.50 | 15.57 | 13.76 |
| | 100.00 | 100.00 | 100.00 | 100.00 |
| Dew Point of Combustion Gas, °F | 135 | 135 | 129 | 124 |

Bases
1) One million Btu of net calorific heat input.
2) Air at 40°F wet bulb temperature compressed and supplied to turbine combustor at 600°F.
3) All fuel gases enter combustor at 77°F.
4) Combustion occurs at 140 psig.
5) Fuel gases (except for natural gas) expected to contain 0.3% water.

## Table 5 (Continued)

| | High Btu Natural Gas | Bi-Gas | Air-Blown Fixed Bed | K-T Utility Gas |
|---|---|---|---|---|
| DATA ON COMBUSTION AT FIXED MAXIMUM FIRING TEMPERATURE OF 1950°F | | | | |
| Lb-Moles Wet Air Required | 86.47 | 80.84 | 71.49 | 81.36 |
| Lb-Moles Theoretical Combustion Gas | 89.05 | 89.85 | 84.14 | 86.75 |
| Lb. of Theoretical Combustion Gas | 2532 | 2498 | 2422 | 2532 |
| Percent Excess Air | 213 | 246 | 206 | 288 |
| Composition of Complete Combustion Gas, Vol% | | | | |
| $CO_2$ | 3.35 | 6.12 | 6.61 | 6.99 |
| $N_2$ + Argon | 75.68 | 72.92 | 73.89 | 73.39 |
| $H_2O$ | 7.28 | 7.19 | 7.62 | 5.16 |
| $O_2$ | 13.69 | 13.77 | 11.88 | 14.46 |
| | 100.00 | 100.00 | 100.00 | 100.00 |
| Dew Point of Combustion Gas, °F | 100 | 99 | 101 | 88 |
| DATA ON COMBUSTION AT FIXED MAXIMUM FIRING TEMPERATURE OF 2400°F | | | | |
| Lb-Moles Wet Air Required | 62.02 | 56.67 | 47.95 | 57.19 |
| Lb-Moles Theoretical Combustion Gas | 64.81 | 62.52 | 60.59 | 62.58 |
| Lb. of Theoretical Combustion Gas | 1330 | 1804 | 1745 | 1838 |
| Percent Excess Air | 110 | 142 | 112 | 173 |
| Composition of Complete Combustion Gas, Vol% | | | | |
| $CO_2$ | 4.61 | 8.48 | 9.18 | 9.69 |
| $N_2$ + Argon | 74.78 | 70.92 | 72.25 | 71.56 |
| $H_2O$ | 9.60 | 9.54 | 10.14 | 6.73 |
| $O_2$ | 11.01 | 11.06 | 8.43 | 12.02 |
| | 100.00 | 100.00 | 100.00 | 100.00 |
| Dew Point of Combustion Gas, °F | 110 | 110 | 113 | 110 |
| DATA ON COMBUSTION AT LEAN FLAMMABILITY LIMITS | | | | |
| Percent Fuel in Air-Fuel Mixture | 5.52 | 7.31 | 12.32 | 7.74 |
| Percent Excess Air | 86 | 388 | 385 | 453 |
| Theoretical Flame Temp, °F | 2710 | 1605 | 1540 | 1595 |

zero percent overall excess air, no derating of conventional boiler equipment is expected for the K-T utility gas due to increased mass velocity which is proportional to draft loss. Likewise, reduced quantities of combustion gas result in less loss of sensible heat whenever combustion gas leaves waste heat recovery equipment. The gas from the airblown process, on the other hand, results in about 25 percent more combustion gas than that which results from natural gas combustion. Dew point of the K-T theoretical combustion gas (124°F) is slightly lower than that resulting for theoretical combustion of the other gases used in the comparison (129° to 135°F), so that no problems are expected with operation of waste heat recovery equipment with the K-T gas. Combustion gas, resulting from K-T utility gas, contains a higher concentration of $CO_2$ than that from the other gases (23.2 percent versus 9.9 to 18.3 percent), resulting in increased emissivity of the gas. This is advantageous in the operation of boiler radiation sections.

Gas turbines operate in practice with excess air to insure complete combustion as well as to control firing temperatures. Table 5 presents a comparison of the K-T gas versus other gases wherein the maximum theoretical firing temperature is controlled at 1,950°F–typical for firing temperature of present generation gas turbines and at a firing temperature of 2,400°F, which has been projected by turbine manufacturers for advanced generation machines. As can be seen, the K-T gas results in no sacrifice of the all-important mass flow through the gas turbine. Most of the combustion gas weight results from air which must be compressed and supplied to the turbine combustor. Air constitutes a higher proportion of this weight, (97-98 percent) in the case of natural gas, than that which occurs in fuels derived from coal gasification.

Recently, much attention has been focused on the control of oxides of nitrogen from combustion sources. Formation of these oxides is influenced by combustion temperature; (a 200°F increase in temperature can more than tripple the rate of $NO_x$ formation); hence, a number of successful techniques are being used on boilers and gas turbines to control the maximum combustion temperature. These methods include water injection into gas turbines and stage-firing techniques on boilers. Much of the recent development work is concentrated on so-called "dry fix" methods which employ burners designed to obtain rapid and effective mixing of the fuel with air to maintain low temperature and short residence time at the burners. Combustion methods designed for $NO_x$ abatement often present secondary disadvantages. For instance, field work is being done by some investigators to control $NO_x$ by combustion with less than theoretical air (off-stoichiometric firing) in order to reduce both the temperature and the concentration of free oxygen in the combustion gas. Unfortunately, this process leads to problems with corrosion or with incomplete combustion and unwanted side products such as ammonia. Therefore, the success of $NO_x$ abatement technolgoy is difficult to predict without extensive field work or without thorough attention to design details.

The K-T gas affords a higher maximum flame temperature (4,280°F) than that which occurs for the other gases used in the comparison (figures 6 and 7); thus, the K-T gas is well-suited for high temperature applications. However, in the case of gas turbines, firing temperatures are prescribed by the design of critical hot gas components of the turbine. On other combustion equipment, high temperature may be prohibited by $NO_x$ formation, in which case, excess air is merely added.

The K-T utility gas offers important properties not available with natural gas, insofar as design of $NO_x$ control systems are concerned. In order to sustain combustion at the burner, the fuel-air mixture must be within the explosive limits, which is an important consideration in design of premix burners commonly used on turbines. Table 5 compares the combustion characteristics of other gases with those of the K-T utility gas at the lean explosive limits. As can be seen, if more than 86 percent excess air is added to natural gas at the burner, the fuel-air mixture will not ignite. At this level of excess air, the theoretical flame temperature is greater than 2,700°F. Therefore, when firing temperatures are to be controlled, it is the practice with some combustors operating on natural gas to use dilution holes (or alternate aerodynamic methods), to add excess air between the burner and the entry to the turbine nozzles. These dilution holes are placed as close to the burner as possible to minimize the time at which the gas is exposed to high temperature without extinguishing the flame. The K-T utility gas, however, has such a low lean explosive limit that up to 453 percent excess air may be added at the burner, resulting in a flame temperature of about 1,600°F, which is still above the auto-ignition temperature of the gas. Hence, for firing temperatures in the range of 1,950-2,400°F, all of the required excess air could be added directly at the burner in the case of the K-T gas, and no "peak" temperature is experienced. Peak flame temperatures can also be controlled by maintaining fuel-rich conditions at the burner, e.g., in stage-firing of boilers. The K-T gas again offers an analagous

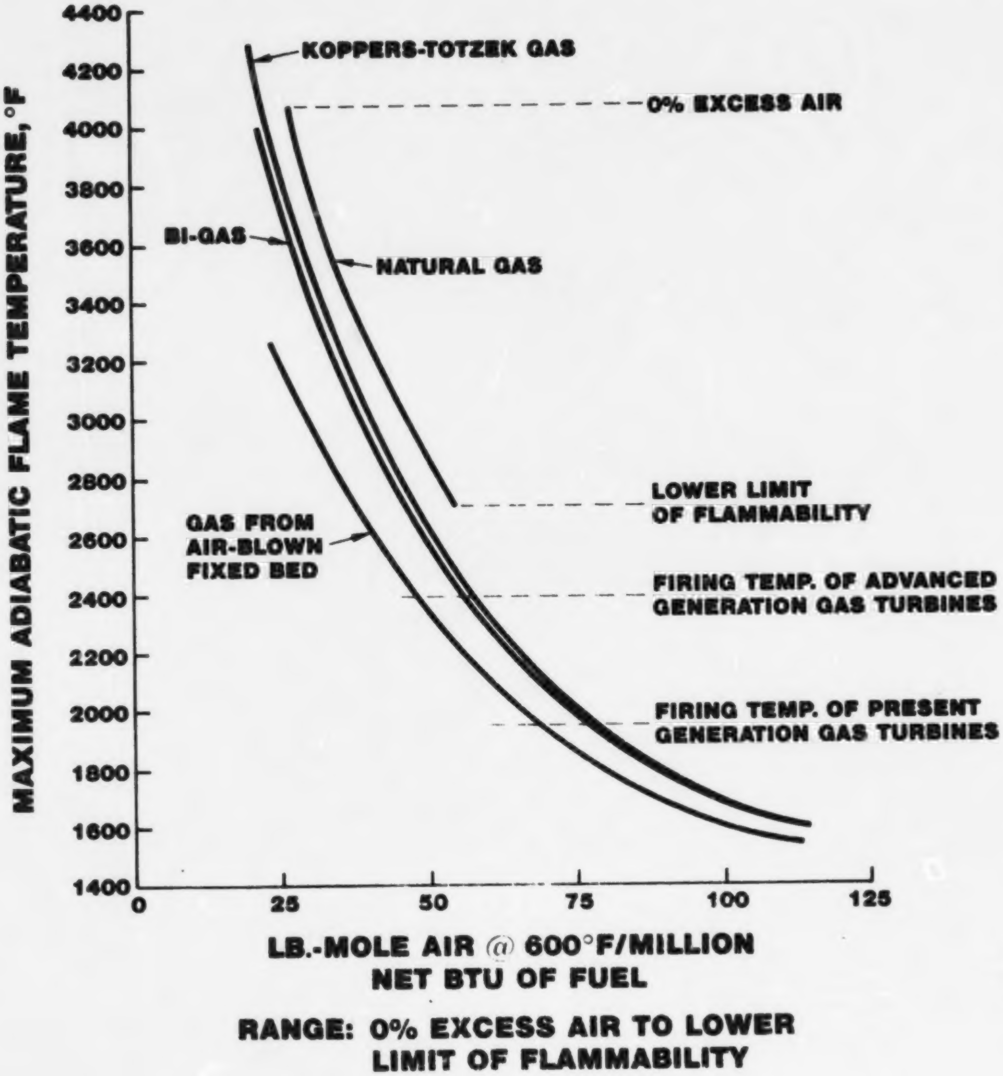
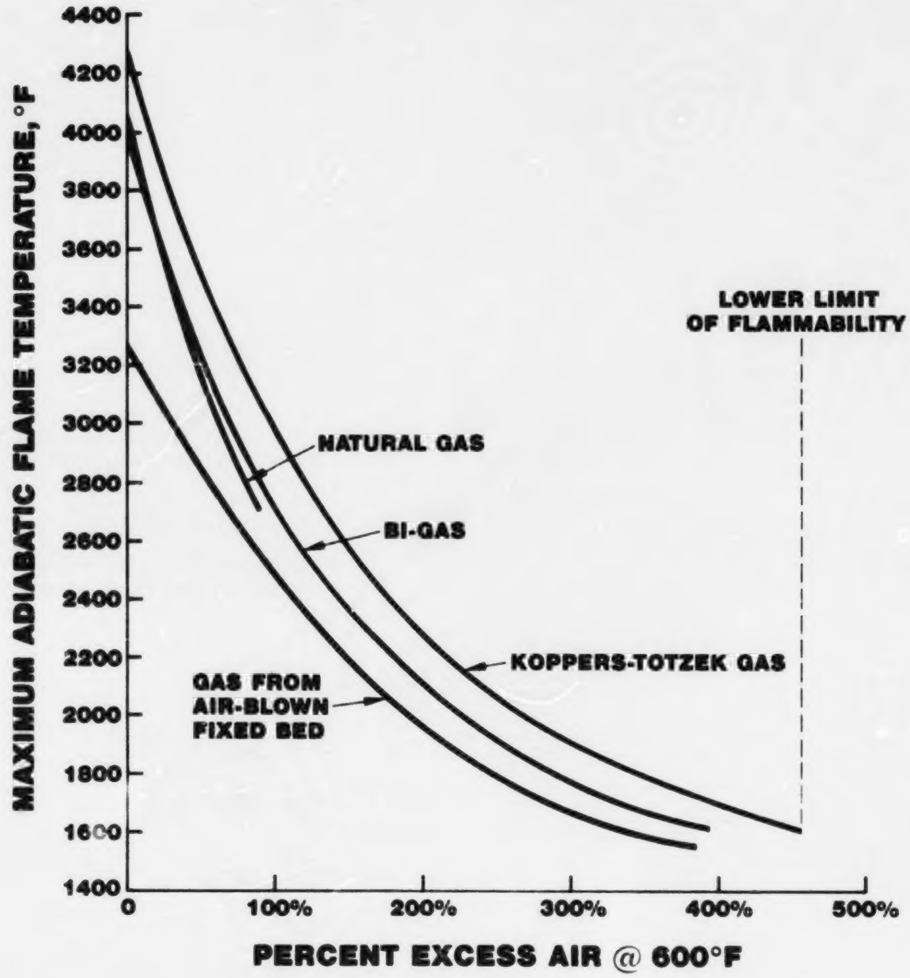


FIGURE 6. FLAME TEMPERATURE VERSUS AIR USED FOR COMBUSTION

advantage to natural gas in this instance, since the rich flammability limit of the K-T gas is much higher than the rich flammability limit of natural gas (75 percent versus 15 percent fuel in fuel-air mixture). A fuel such as the K-T gas, with a high ratio of rich-to-lean flammability limit, will display excellent combustor performance.

Simplicity, controlability and reliability of the K-T process are important factors in the operation of a clean and environmentally acceptable gasification plant. As reported in the pollution evaluation report, K-T plants can be designed to meet current environmental codes. We in Koppers believe that these codes can be attained with current technology and equipment at economic costs.

# FLAME TEMPERATURE VERSUS EXCESS AIR USED FOR COMBUSTION



**RANGE: 0% EXCESS AIR TO LOWER LIMIT OF FLAMMABILITY**

FIGURE 7

# ENVIRONMENTAL ASPECTS OF THE BI-GAS PROCESS

R. J. Grace and E. K. Diehl*

*Abstract*

*Because of the basic nature of the BI-GAS process, control of those elements of the process that might potentially affect the environment is believed to be well within the realm of current, known technology.*

*Coal preparation is essentially no different than that which is considered common practice in the power industry today. Reactive components in the coal are completely used in the process, and the ash for disposal is in the form of granular, vitreous slag. Sulfur appearing in the feed coal is ultimately transformed in the process to its elemental form by way of the gas cleanup system and its subsequent pass through a conventional Claus plant.*

*The severity of the gasification step results in minimum formation of those compounds that are normally considered potential water pollutants.*

*Thus, environmental aspects of the BI-GAS process center more on those factors that are associated with the production of coal, handling of slag, and supply of process water, rather than on anything unique in the process itself.*

## INTRODUCTION

It is obvious that any new development such as the conversion of coal to supplement natural gas supplies will have some unavoidable effect on the environment. Readily identifiable is the prospect of an expanded coal mining industry to supply the raw material for the large gasification plants that are projected as being necessary during the next several decades. Many of these plants will be located on sites adjacent to coal deposits that can best be won by stripping operations. Thus, ecological effects of such operations will represent an important facet of the new coal-to-gas industry. Adherence to good mining practices, plus the promise of new developments in the technology of mined-land reclamation, should serve to minimize any permanent, irreversible impact upon the environment.

The total water requirements for commercial† coal gasification plants are estimated to be in the range of 27 to 32 million gallons per day, depending upon the rank of the coal gasified. If partial air cooling is employed, water requirements can be reduced to between 14 and 16 million gallons per day. In either case, a portion of the water supplied, approximately 2.5 million gallons per day, is consumed in the process to provide hydrogen for gasification. Water availability and water management, therefore, are other areas where environmental compatibility will have to be considered.

Another and often overlooked ecological effect is the "people factor." For example, a commercial coal gasification plant would employ approximately 370 persons. This group would include operating personnel, maintenance crew, and the associated supervisory, technical, and clerical personnel. Additional manpower would be required for other steps in the total operations. Depending upon the specific gasification process, power plant requirements could add as many as 230 persons to the total plant force.

Separate from the coal gasification plant itself is the manpower required to supply the coal. Assuming that there is a plant production requirement of 15,000 tons per day, the mining workforce would range from 434 men for strip mine operation to 1,340 for deep mine operation. These estimates are based on recent productivity figures (ref. 1) of 11.2 tons per man per day for underground mines and 34.6 tons per man per day for strip mines.

It is conceivable, then, that a commercial coal gasification installation, including coal production facilities and depending upon its location, could develop a manpower requirement of as many as 1,940 employees. Assuming that each employee represents a family of four, nearly 8,000 people would be related directly to the plant. Since most plant sites would be near the coal, and therefore not near existing towns, it is probable that new communities would need to be established. Already, new communities are being designed to handle the expected influx of people. Though only indirectly related to the environmental impact of coal gasification technology, the ecological effects of the "people factor" would demand serious consideration.

Each of the above is a part of the environmental

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†For purposes of projections and conceptual design studies, a commercial plant is defined as having the capacity to produce 250 million standard cubic feet per day of SNG.

impact of all gasification processes. The problems created by each are receiving the attention of knowledgeable experts and are separate and apart from the environmental aspects of the technical process.

### *The BI-GAS Process*

It should be emphasized at the outset that specific details regarding the environmental impact of the BI-GAS process cannot be established factually at this stage in the process development. When the pilot plant, now under construction at Homer City, Pennsylvania, is operational in mid-1975, many of the unit operations will be tested for the first time. Operation of the pilot plant will be in the nature of process research. Experience during operation may well bring about some changes in the process that could influence design of the subsequent commercial plant.

Nevertheless, certain features of the BI-GAS process can be assessed in relationship to their influence on the environmental compatibility of the process. The heart of the BI-GAS process is the two-stage gasifier which uses coal in entrained flow. (See fig. 1.) Fresh pulverized coal will be introduced into the upper section (Stage 2) of the gasifier at pressures in the range of 70 to 100 atm. Here, the coal will come in contact with a rising stream of hot synthesis gas produced in the lower section (Stage 1) and be partially converted into methane and more synthesis gas.

The residual char entrained in the raw product gas will be swept upward and out of the gasifier. The char will be separtated from the product gas stream and recycled to the lower section (Stage 1) of the gasifier.

In the lower section, the char will be completely gasified under slagging conditions with oxygen and steam, producing both synthesis gas and the heat required in the upper section (Stage 2) for partial gasification of the fresh coal.

The raw product gas from Stage 2 will be shifted to adjust the hydrogen to carbon monoxide ratio, purified by removal of hydrogen sulfide and carbon dioxide, and upgraded in Btu content to pipeline quality by catalytic methanation.

From the standpoint of environmental consideration, several advantages of the BI-GAS concept are:

1. Because it employs an entrained rather than a fixed- or fluidized-bed system, all types of coal should be amenable without special preparation or prior pretreatment;
2. All feed coal is consumed in the process; and
3. Principal byproducts are slag for disposal and elemental sulfur.

The actual daily coal requirement for the BI-GAS process will vary somewhat, depending upon the rank and source of the coal. The quantities presented in this paper are based upon the pilot plant design and reflect the use of Pittsburgh seam coal containing 7.03 percent ash and 2.46 percent sulfur.

### *Coal Preparation*

Using Pittsburgh seam coal, a commerical BI-GAS plant would gasify about 12,000 tons per day. An estimated additional 1,700 tons per day would be required for auxiliary purposes such as for steam generation and drying. Thus, the daily coal requirement for the process would be some 13,700 tons per day of cleaned coal.

Assuming a 25 percent refuse loss from cleaning, 18,267 tons per day of run-of-mine coal will be processed in a cleaning plant at either the mine site or at the gasification plant. In either case, conventional cleaning, with the necessary environmental control, can be applied. Refuse for disposal will amount to 4,567 tons per day.

Further coal preparation consists only of reducing the size of the cleaning plant product (1½ inch X 0) to the size required for the process; namely, 70 percent minus 200 mesh. The grinding and pulverizing will be done in two stages. The coal is first reduced in a cage mill. The cage mill product is pulped with water, and final pulverization is accomplished in a wet ball mill.

The latter stage, wet pulverization, is incorporated into the process as a step in the subsequent high pressure slurry feed system. Its use, however, will eliminate some of the potential dust problems that are associated with dry pulverization and with transport of dry pulverized coal.

### *Slag*

As mentioned earlier, 12,000 tons of coal per day will be completely consumed in gasification. The slag produced will amount to 844 tons per day.

In the BI-GAS gasifier, molten slag from Stage 1 will drop into a reservoir of water for rapid quenching, thus causing the slag to shatter into small pieces. The resulting granules of vitreous slag will be removed to a settling pond. Clarified water from the settling pond will be recycled, and the slag will be removed for disposal.

While the exact nature of the slag produced from a

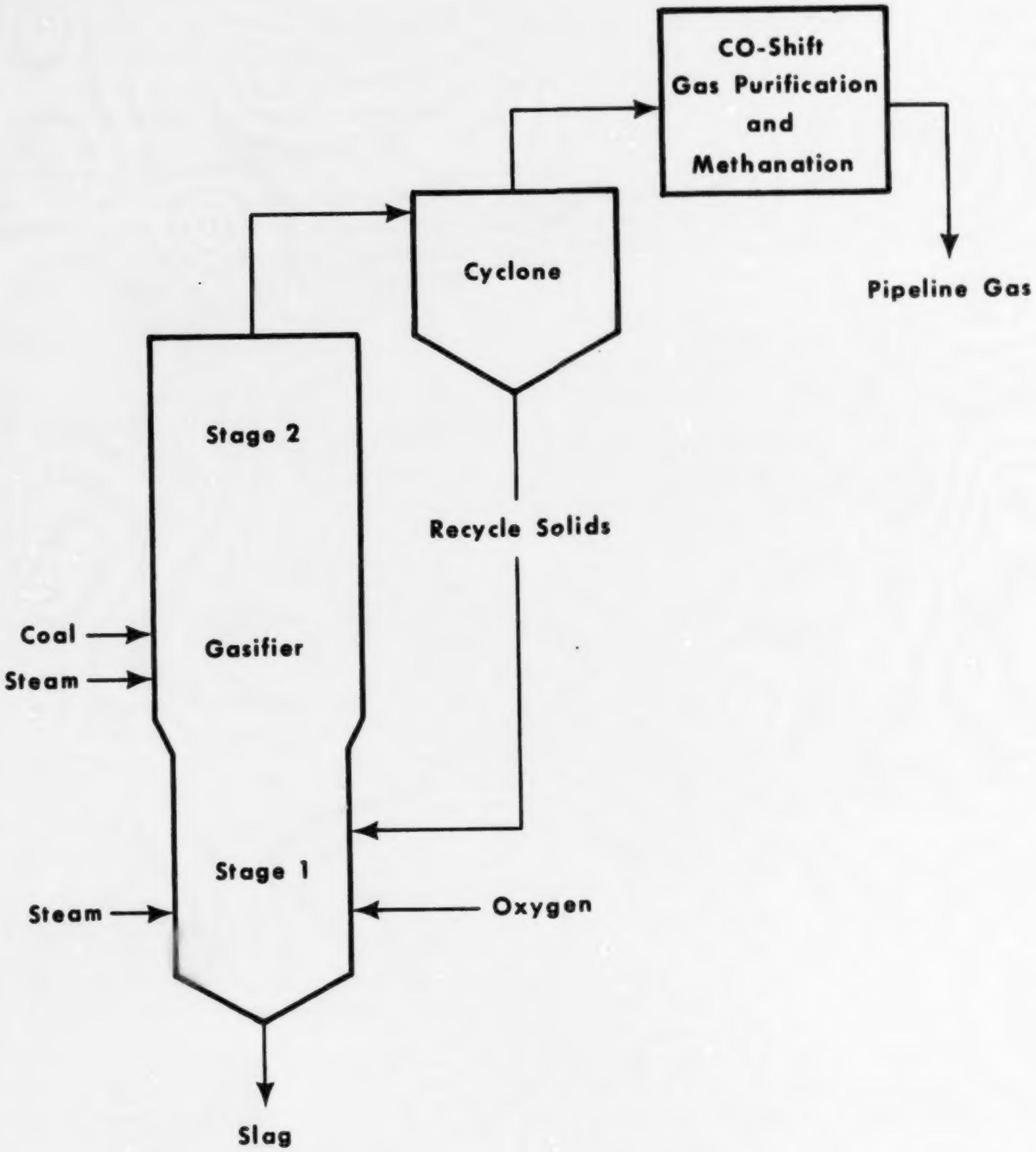
CO-Shift
Gas Purification
and
Methanation
Cyclone
Pipeline Gas
Stage 2
Recycle Solids
Coal
Steam
Gasifier
Stage 1
Steam
Oxygen
Slag

Figure 1. Simplified Flow Diagram for the BI-GAS Process

particular coal under BI-GAS Stage 1 conditions is yet to be determined, it is not expected to be too unlike that which results from conventional slag-tap boiler operation. The vitreous nature of the slag should minimize concern over water-leaching in the event that the material is returned to the mine site or is used elsewhere as land fill. It is possible that a productive use of the slag may be found that would change a process waste into a useful material.

*Sulfur*

About 95 percent of the sulfur entering with the coal is expected to leave the gasifier as hydrogen sulfide. The remaining sulfur will leave in compounds such as carbonyl sulfide and carbon disulfide. The sulfur in these compounds will be hydrogenated into hydrogen sulfide after passing through the catalyst guard filters upstream of the shift reactor. As a consequence, it appears that all of the sulfur in the gas will enter the acid gas removal facilities as hydrogen sulfide.

Hydrogen sulfide will be removed from the gas stream in a SELEXOL unit and converted to elemental sulfur in a Claus unit. Both of these steps presently meet environmental standards.

About 295 tons of sulfur per day will be produced in the commercial BI-GAS plant. In its elemental form, it will be the most compatible with environmental requirements.

*Process Water Contaminants*

Because of the conditions existing within the gasifier, the BI-GAS process is not expected to produce oils, tars, or phenols, which in some processes represent an important contaminant in process water streams. None of these were detected during the extensive experimental work that preceded design of the pilot plant. Their absence will be confirmed when the pilot plant goes into operation.

The major process water contaminant will be ammonia. The ammonia content of the gas stream is not known at this time. It has been estimated that about 70 percent of the nitrogen in the feed coal will form ammonia in most SNG processes. Based on that assumption, the BI-GAS process would produce about 123 tons per day. Most of this would appear in the water from the gas scrubber and in the shift condensate. Conventional waste water treatment methods can adequately remove ammonia, which may be of sufficient purity for commerical use.

## SUMMARY

The information presented should be considered as the principal environmental aspects quantifiable at this time. At its present stage of development, BI-GAS promises some distinct advantages with regard to its environmental compatibility. Confirmation of these advantages will necessarily have to await operation of the total process at the pilot plant scale.

Process characterisitics that are unique to BI-GAS appear to favor control of some elements of the overall environmental impact that will accompany a coal gasification industry. Those problems that are common to all processes can be solved, to a great extent, with existing commerical technology and good engineering practice.

## ACKNOWLEDGMENT

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# SULFUR EMISSION CONTROL WITH LIMESTONE/DOLOMITE IN ADVANCED FOSSIL FUEL-PROCESSING SYSTEMS

**Dale L. Keairns, Eoin P. O'Neill, David H. Archer***

*Abstract*

*High temperature sulfur removal can be achieved with limestone and/or dolomite influidized bed fuel processing systems, now being developed for power generation. Westinghouse is developing this process for low-Btu gasification and fluidized bed combustion of fossil fuels. The use of limestone/dolomite for high temperature emission control offers the opportunity to meet sulfur dioxide emission standards; to minimize the environmental impact of spent solids; and to permit greater simplicity and versatility, increased plant efficiency, and reduced electrical energy costs over competitive processes.*

*Alternative sulfur removal system concepts have been experimentally studied for each fuel-processing system: once-through and regenerative sorbent operation, pressurized and atmospheric pressure operation, spent stone processing, and disposal/utilization of processed stone. A thermogravimetric analysis system, capable of operating at elevated pressure with corrosive gases, is being used to collect kinetic data over a wide range of operating conditions--e.g., stone type, temperature, pressure, particle size, and gas composition. Illustrative data on sulfur removal and regeneration are present for each fuel processing system and are being used for establishing design and operating conditions.*

## INTRODUCTION

Westinghouse is working on a multifaceted program to develop advanced coal, oil, and low-grade fuel processing for electric power generation (refs. 1,2). Fluidized bed combustion and low-Btu gasification processes for fossil fuels are being developed as shown in figure 1. The fluidized bed coal-gasification process will be studied in a 1,200 lb coal/hr process development unit which will be operational in mid-1974. A commercial demonstration plant design has been initiated. Commercial power plant designs for pressurized fluidized bed combustion systems have been completed (ref. 1). Laboratory and pilot plant tests have been and continue to be carried out by several contractors as part of the EPA and OCR program. A preliminary design for a 30 MW pressurized fluidized bed combustion boiler development plant has been completed. Preliminary design studies have been carried out for the pressurized oil gasification system. Fluidized bed oil gasification at atmospheric pressure has been studied by Esso (U.K.) in a 1 MW pilot plant (ref. 4). A 50 MW demonstration plant design is underway (ref. 1).

These processes offer a solution to the problem posed by the apparently conflicting and urgent requirements of efficient power generation and pollution abatement. Each of these processes has the potential to reduce power plant capital costs; increase overall operating efficiency; minimize emissions of $SO_2$, $NO_X$, and particulates; and avoid the generation of other environmental problems. The technologies involved in the four processes have fundamental similarities as illustrated in figure 1. One similarity is the use of limestones and/or dolomites for high temperature sulfur removal during the fuel-processing step.

The sulfur removal system concepts explored utilizing limestone and/or dolomite are illustrated in figure 2. The sulfur is removed in solid form as calcium sulfate or calcium sulfide with greater than 90 percent reduction in sulfur emission (ref. 5). The spent stone may be regenerated to produce a reusable sorbent for sulfur removal. The sulfur-rich gas released as sulfur dioxide or hydrogen sulfide during the regeneration process could be sent to a sulfur or sulfuric acid recovery plant. This provides the opportunity to minimize the sorbent feed requirements and the spent stone. The molar feed requirement of calcium to remove each mole of sulfur in the fuel may be significantly less than one, the stoichiometric ratio, for this regeneration process option. Regeneration of the sorbent to minimize the stone requirements is desirable; however, sulfur recovery may not be desirable or economically attractive. An alternative regeneration process concept can be employed in specific cases which does not require sulfur recovery. The sulfur-rich gas from the regeneration process can be stoichiometrically

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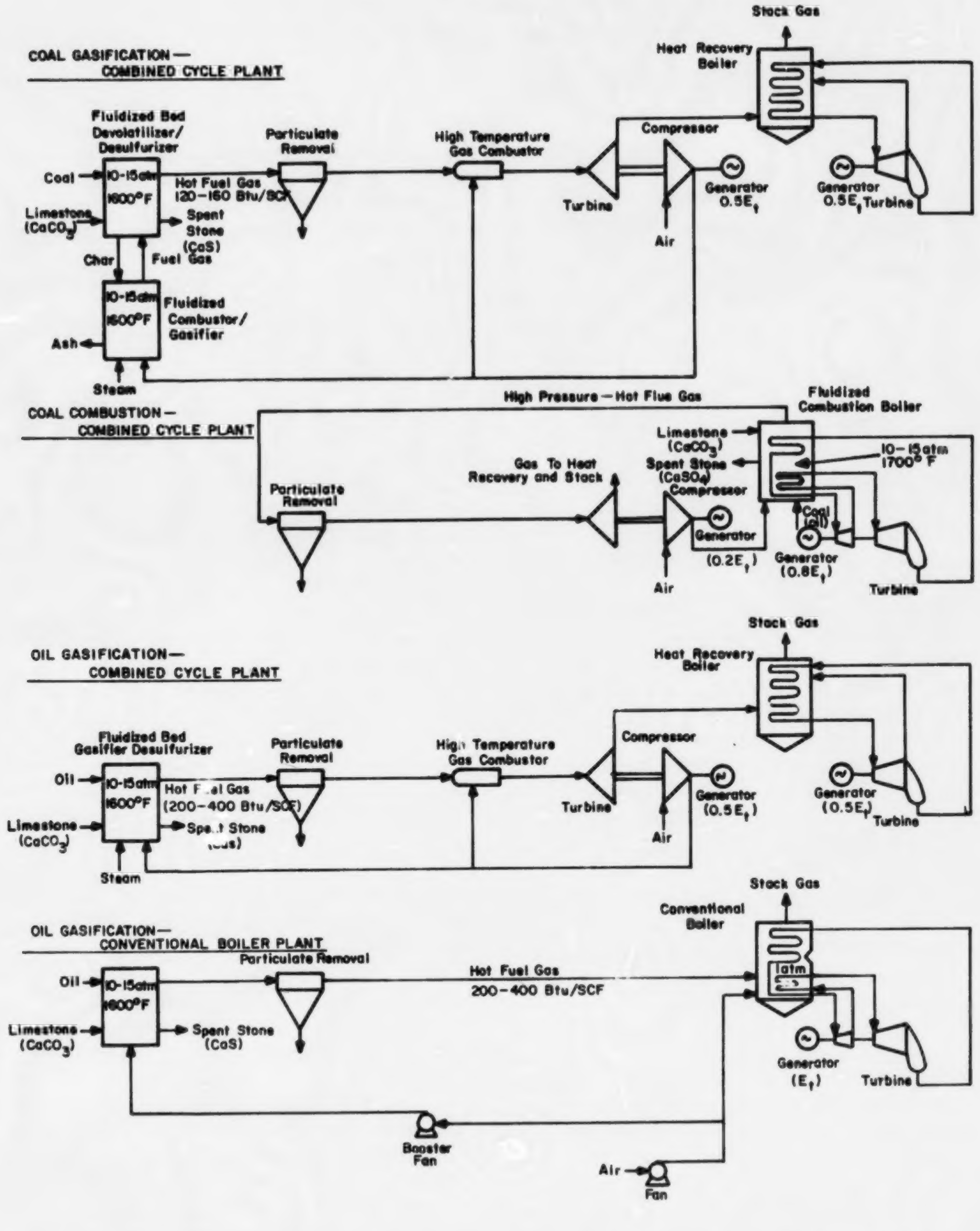
COAL GASIFICATION— COMBINED CYCLE PLANT
Stack Gas
Heat Recovery Boiler
Fluidized Bed Devolatilizer/ Desulfurizer
Particulate Removal
High Temperature Gas Combustor
Compressor
Coal
10-15atm
1600°F
Hot Fuel Gas 120-160 Btu/SCF
Limestone (CaCO3)
Spent Stone (CaS)
Char
Fuel Gas
Turbine
Generator 0.5Et
Generator 0.5Et Turbine
Air
10-15atm
1600°F
Fluidized Combustor/ Gasifier
Ash
Steam
High Pressure — Hot Flue Gas
COAL COMBUSTION— COMBINED CYCLE PLANT
Fluidized Combustion Boiler
Limestone (CaCO3)
Spent Stone (CaSO4)
10-15atm 1700° F
Gas To Heat Recovery and Stack
Compressor
Particulate Removal
Coal (oil)
Generator
(0.2Et)
Generator (0.8Et)
Air
Turbine
OIL GASIFICATION— COMBINED CYCLE PLANT
Stack Gas
Heat Recovery Boiler
Fluidized Bed Gasifier Desulfurizer
Particulate Removal
High Temperature Gas Combustor
Compressor
Oil
10-15atm
1600°F
Hot Fuel Gas (200-400 Btu/SCF)
Limestone (CaCO3)
Spent Stone (CaS)
Turbine
Generator (0.5Et)
Generator (0.5Et)
Turbine
Air
Steam
OIL GASIFICATION— CONVENTIONAL BOILER PLANT
Stack Gas
Conventional Boiler
Particulate Removal
Oil
10-15atm
1600°F
Hot Fuel Gas 200-400 Btu/SCF
1atm
Limestone (CaCO3)
Spent Stone (CaS)
Generator (Et)
Turbine
Booster Fan
Air
Fan

FIG. I. FLUIDIZED BED FUEL PROCESSING FOR POWER GENERATION FROM FOSSIL FUELS

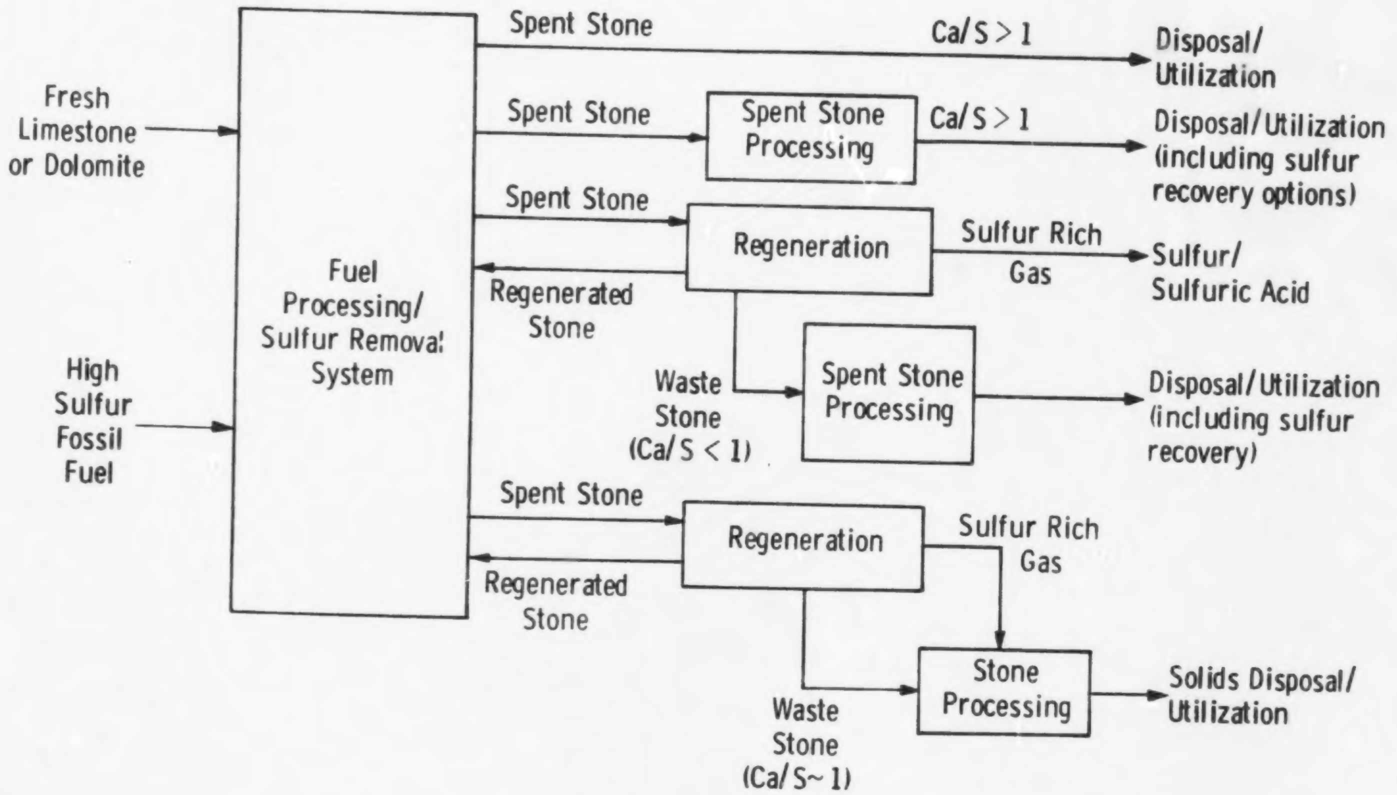
Fresh Limestone or Dolomite
High Sulfur Fossil Fuel
Fuel Processing/ Sulfur Removal System
Spent Stone
Ca/S > 1
Disposal/ Utilization
Spent Stone
Spent Stone Processing
Ca/S > 1
Disposal/Utilization (including sulfur recovery options)
Spent Stone
Regeneration
Regenerated Stone
Sulfur Rich Gas
Sulfur/ Sulfuric Acid
Waste Stone (Ca/S < 1)
Spent Stone Processing
Disposal/Utilization (including sulfur recovery)
Spent Stone
Regeneration
Regenerated Stone
Sulfur Rich Gas
Waste Stone (Ca/S~ 1)
Stone Processing
Solids Disposal/ Utilization

Figure 2— Sulfur removal system process concepts

reacted with a side stream of stone from the regenerator to produce calcium sulfate in a separate vessel. This concept provides the potential to maintain the stone requirement near stoichiometric, to produce a solid suitable for disposal, and to eliminate the requirement for sulfur recovery. The sulfur removal system may also be operated as a once-through sorbent system. The spent stone may be processed for disposal without regeneration or may be suitable for direct disposal as in the fluid bed combustion system when operated to produce calcium sulfate/calcium carbonate. The once-through option results in a greater stone requirement and higher operating cost which must be balanced against the potential for reduced capital cost, reduced space requirements, increased plant efficiency, and improved reliability.

### *Experimental Program*

Laboratory support studies are being carried out at Westinghouse to demonstrate the alternative steps in the sulfur removal system concepts (ref. 6). The overall object of the study is to determine the optimum operating conditions for sulfur removal systems within the engineering restraints imposed by the energy-recovery objectives of the system.

## APPARATUS

The primary experimental study is centered on the use of a specially modified thermogravimetric apparatus (TG). The TG is an electrically recording balance which measures the changes in weight of a solid suspended from the balance arm in an electronically controlled furnace. The gaseous stream flowing over the solid and the temperature of the furnace may be adjusted or altered as desired, within specifiable limits. The capability to study gas/solid reactions at pressures up to 30 atmospheres at temperatures up to 1,200°C and provision for the use of corrosive gases at pressure required modifications to the commercial TG systems which have been described elsewhere (ref. 1). The primary data are the rates of the gas-solid reactions under the experimental conditions of sorbent pretreatment, temperature, pressure, gas composition, sample geometry, gas flow in the reactor, and the extent of reaction, or utilization of the sorbent.

With this equipment, it is possible to measure limiting rates of reaction under conditions where properties internal to the sorbent prevent efficient application of the reactions. It also provides a measure of the capacity of the sorbent samples for reaction (extent to which the solid reacts).

## SCOPE OF EXPERIMENTAL PROGRAM

Fundamental data are required to determine the feasibility and optimum conditions for each processing step in each sulfur removal system, i.e., removal, regeneration, spent stone processing, and confirmation of inactivity of the waste stone for disposal. Thermogravimetric studies pertinent to each of these areas have so far involved over 300 experiments on the reactions of limestone, dolomites, and their chemical derivatives. These experiments include routine tests for stone selection, sulfation and cyclic sulfidation-sorbent regeneration experiments, feasibility tests on variations of recarbonation or resulfation processes for dry spent stone disposal, and activity tests on processed stone for disposal. Illustrative examples of results and conclusions in the areas of sulfur removal and sorbent regeneration are presented in this paper to demonstrate the contribution such a study makes to the overall objectives of developing commercial sulfur removal systems for advanced fossil-fuel-processing systems. The results of this work have been and are being used as a basis for sulfur removal system design and economic studies, for both fluidized bed combustion and fluidized bed gasification processes.

## SORBENT FORM

Gasification or combustion in a pressurized system (10 or 15 atmospheres) utilize combined gas and steam turbines for power generation which can provide increased overall plant efficiency. The immediate impact of the pressurized options on the sulfur removal process is to necessitate operation under conditions where calcium carbonate rather than calcium oxide is the stable form of the dolomite/limestone sorbent.

In figure 3, the proposed operating conditions for the pressurized fluidized bed coal gasification system (ref. 2) are shown to favor stability of calcium carbonate rather than calcium oxide. In figure 4, proposed operating conditions for two pressurized fluidized bed combustion systems lie in the region of pressure and temperature where either the carbonate or oxide form of the dolomite is stable. The sulfidation and sulfation reactions of both fully-calcined and half-calcined dolomites have been studied at atmospheric pressure and at 10 atmospheres pressure in order to anticipate the widest range of operating conditions.

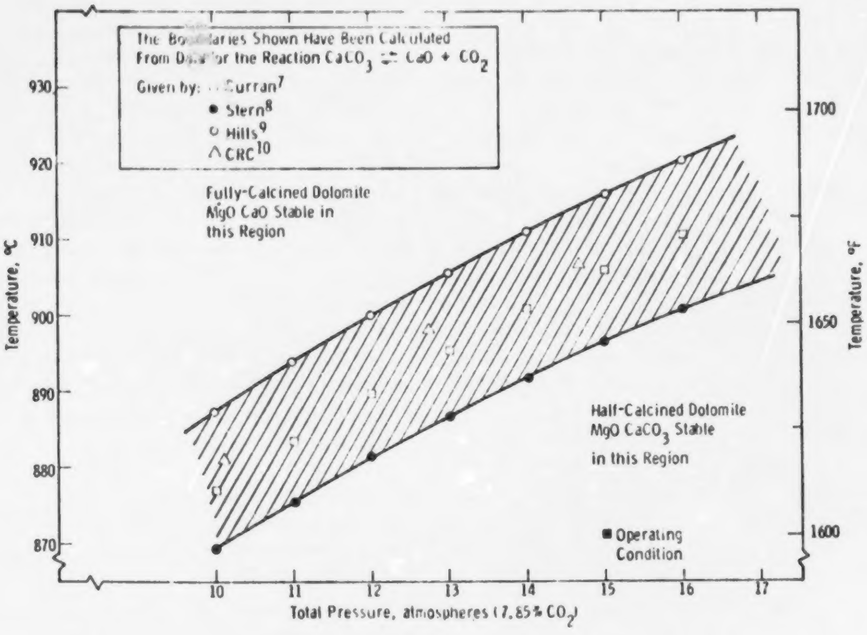

Figure 3 – Temperature and pressure conditions for stability of the sorbent as half-calcined dolomite at the projected desulfurizer outlet composition of 7.85% $CO_2$

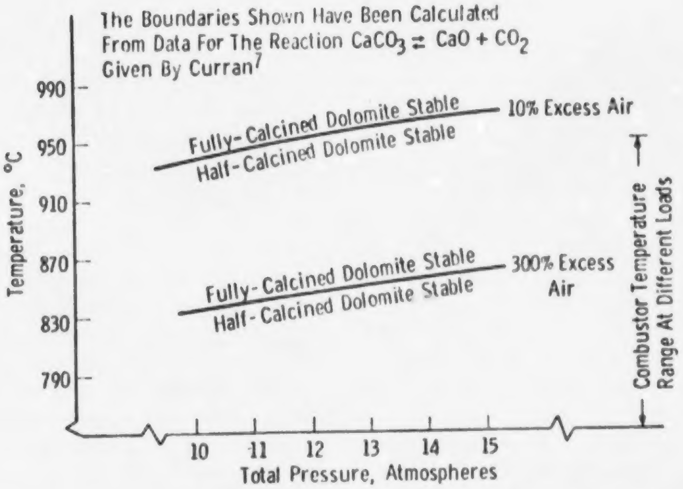

Figure 4– Temperature and pressure conditions for stability of the sulfur sorbent as half-calcined or fully-calcined Dolomite at projected combustor outlet gas compositions

*Sulfur Removal*

The successful demonstration of the primary sulfur capture process has long outdistanced any understanding of the fundamental mechanisms, which limit attainment of thermodynamic yields. While this success might be thought to obviate the need for further investigation, it is clear that the current limits to the flexibility of the processes, and hence to their usefulness, are bounded by physical conditions which cannot be readily optimized since their effects have not been fully explained.

For each of the major fuel-processing options, desulfurization of the product gases has been convincingly demonstrated in fluidized beds, within limiting conditions, by workers at Argonne (ref. 11); Esso (U.K.) (ref 4); Exxon (ref. 12); Consolidation Coal (ref. 13); Pope, Evans, and Robbins (ref. 4); the U.S. Bureau of Mines (ref. 15); and the National Coal Board (U.K.) (ref. 16).

## EFFICIENCY OF SORBENT UTILIZATION IN COMBUSTION SYSTEMS

Sulfur removal in the fluidized bed combustion process is affected by at least 13 variables (ref. 5). Since some of these variables (e.g., temperature, pressure) have a direct bearing on the kinetics of the chemical reactions, their influences have been studied in the TG apparatus and the data so obtained has reproduced the performance of existing fluidized-bed reactors (ref. 17). This ability to correlate TGA data with fluidized bed data has been important in establishing the utility of the TGA.

One important result from the sulfur removal studies under oxidizing conditions relates to the efficiency of use of the sorbent. A substantial body of data shows that Ca/S molar ratios of 2.5 or greater must be maintained in the fluidized-bed in order to achieve 90 percent capture of the $SO_2$ liberated from the fuel. This molar ratio corresponds to a mean sulfation level of 36 percent of the CaO in the fluidized bed. The results obtained with limestone frequently correspond to even lower levels of sulfation at the point where fresh sorbent must be added to retain sulfur removal efficiency.

With dolomite as the sorbent, TG studies have shown that (35 to 40 percent) sulfation is a critical region for the kinetics of sulfation where a transition occurs from a rapid reaction rate to a slow product diffusion-controlled rate. This phenomenon is evident in both the atmospheric pressure and pressurized TG data. The relationship between the reaction rate for sulfation and the type of porosity in the calcined stone has been convincingly demonstrated by Borgwardt (ref. 18). Since the stone pore structure is established as it calcines, this phase of the process offers the possibility of altering the reaction conditions to generate in the stone the desired pore structure. The result of favorably altering the conditions of calcination is shown for the pressurized case in figure 5; the capacity of the stone to react with $SO_2$ in the fast phase of reaction has been doubled. If this process can be duplicated in fluidized-bed reactors, then a Ca/S mole ratio of approximately 1.25 will suffice to meet $SO_2$ emission standards, yielding a more homogeneous product with less residual calcium carbonate or calcium oxide (depends on operating conditions) and halving both the quantities of fresh sorbent required and the waste stone disposal burden.

Pressurized TG experiments show that half-calcined dolomite reacts with $SO_2$/air mixtures at a rate comparable to calcined dolomites (ref. 1). Since the rapid deceleration of rate at approximately 36 percent sulfation noted with calcined stone does not occur with half-calcined dolomite, utilization up to about 60 percent can be projected for this stone. As the decline in rate with sulfur load is not abrupt, increased sorbent efficiency is attainable with modest increases in gas residence time in the bed.

The pressurized fluidized bed combustion process can be operated without heat transfer surface in the combustor by operating with high excess air (ref. 1). Consideration of the adiabatic combustion system using 300 percent excess air as the heat sink to supply power to a gas turbine in a pressurized combustor led to the speculation that the increase in oxygen concentration would increase the sulfur removal efficiency by driving the equilibrium for reaction toward the production of calcium sulfate and by increasing the rate of attainment of equilibrium.

Experiments were carried out on the TG in which the oxygen concentration was increased to 11 percent at 10 atmospheres pressure. Since only minute increases in reaction rate were observed on increasing the oxygen fraction in the sulfating gas, the prospect of enhancing the efficiency of sulfur removal in this fashion can be ruled out (ref. 1). However, increasing the partial pressure of oxygen may be beneficial in extending the temperature range for efficient sulfur removal, indicating that the complexity of interaction of the process variables requires a thorough investigation.

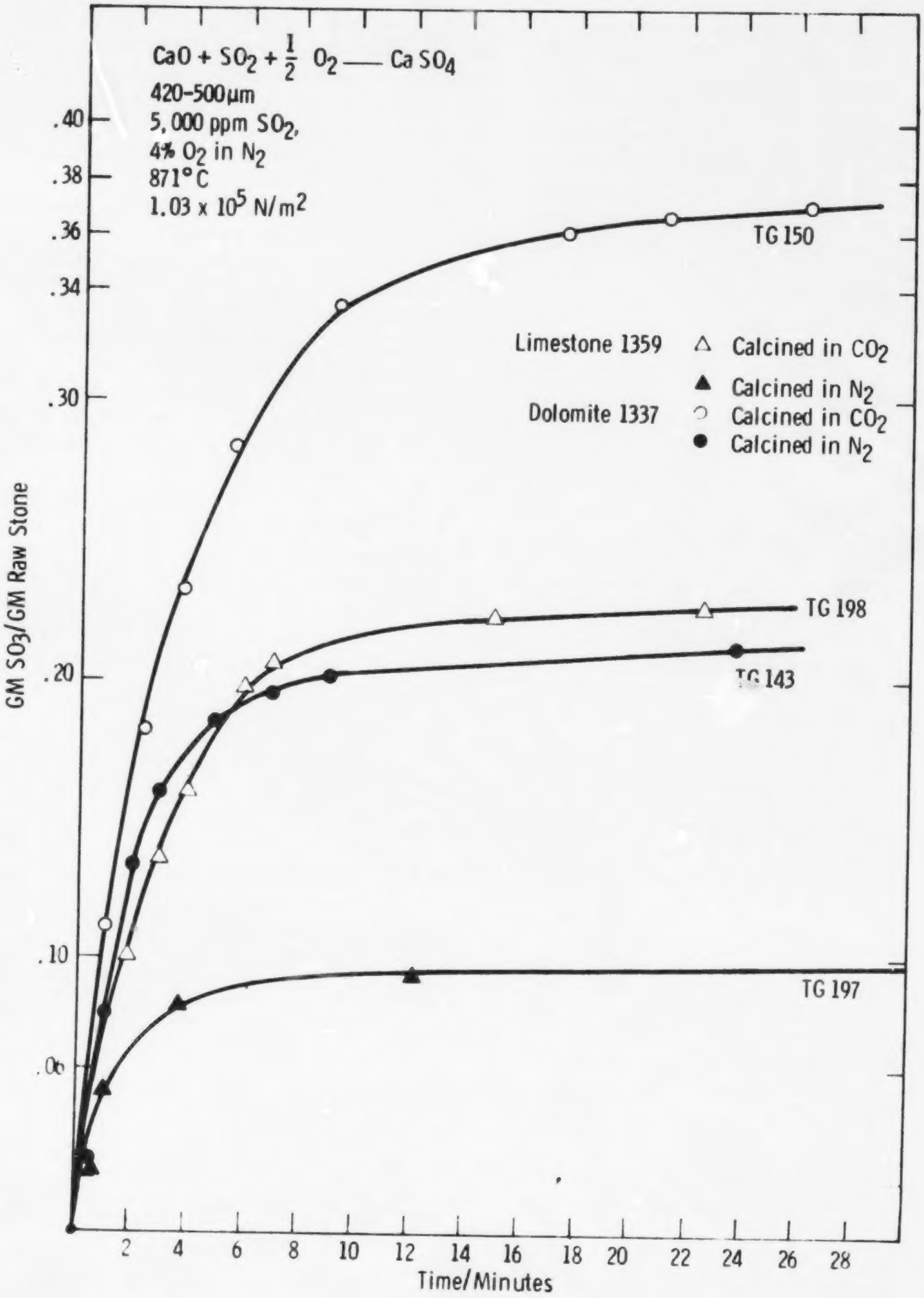

Figure 5—Comparison of pressurized sulfation of limestone and dolomite

## EFFICIENCY OF SORBENT UTILIZATION UNDER REDUCING CONDITIONS

(Coal and Oil Gasification)

For the coal gasification process, TG studies at 10 atmospheres pressure have shown that sulfidation of half-calcined dolomites proceeds rapidly up to greater than 90 percent utilization of the sorbent $CaCO_3$ content, at temperatures in the range 870-930°C and with particle sizes up to 2,000 $\mu$m in diameter. The primary desulfurizing reaction will not limit sulfur removal in this system, and other factors such as stone attrition in the fluidized bed are likely to limit the performance of the process.

The retention of sulfur in fluidized-beds of calcined lime under reducing conditions at atmospheric pressure has been demonstrated by Esso at Abingdon (England) (ref. 4). A perplexing feature of their work has been the decline in sulfur removal activity at low sulfur loading of the sorbent stone (corresponding to about 15 percent utilization of the CaO). The sulfidation of dolomite was shown by Pell (ref. 19) to proceed to completion. Our studies of calcined limestones, ranging from high-purity $CaCO_3$ to dolomites, indicate that little decrease in rate occurs before 30 percent of the CaO is sulfided in 1,000 $\mu$m particles. At higher utilization, the rate of reaction observed on the TG reflects the calcium oxide concentration in the particular limestone (ref. 1). To date, we have not succeeded in explaining the poorer performance of the stones in the pilot plant fluidized-bed oil gasifier. Figure 6 shows the current projection of our data to the physical dimensions of the Esso gasifier.

Possible explanations for the decline in activity which have been advanced, include:

(1) A diffusional barrier to reaction which is created by the carbon which deposits, during gasification, on the sorbent surfaces;

(2) Sintering of the calcined stone at 870°C during its residence in the gasifier bed;

(3) The cycle of oxidizing-reducing conditions to which the stone is exposed causes a buildup of calcium sulfate in the pores of the stone, hindering transport of the sulfiding gases;

(4) The conditions of sulfur removal in the pilot plant, e.g., location of oil injection, bed depth, etc.

The ability to achieve high utilization in the atmospheric-presure oil gasification process would enhance the attractiveness of a once-through sorbent process. Experiments are continuing to develop the potential for high utilization which is evident from the experimental data.

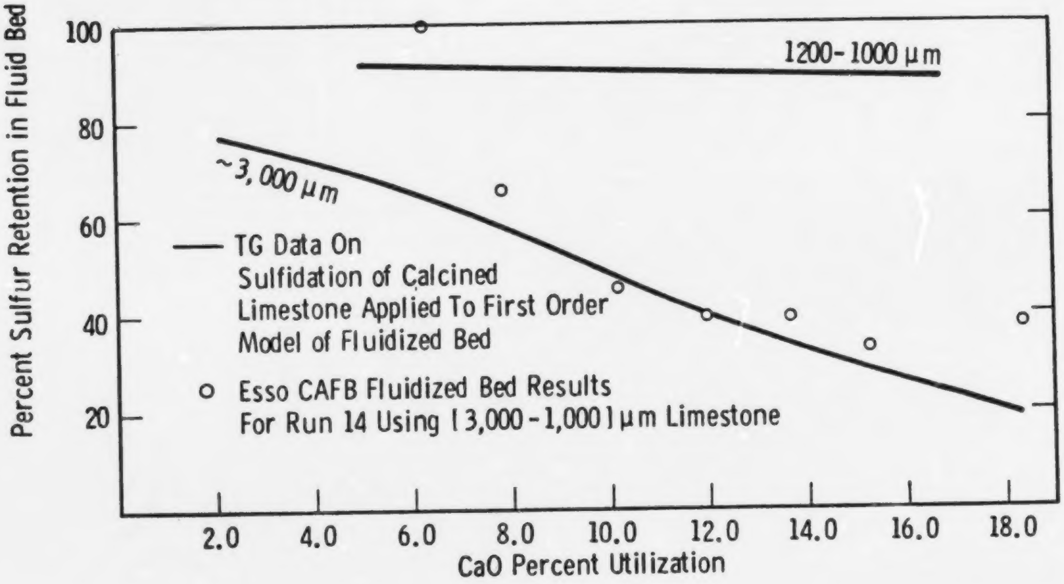

Figure 6–Comparison of Esso CAFB oil gasification desulfurization and the projection of fluidized-bed results from thermogravimetric data

### *Regeneration of Sulfur Sorbent*

The regeneration of the spent sulfur sorbent in a usable form (i.e., with retention of its activity for the sulfur removal reactions) with or without recovery of the sulfur is economically and environmentally desirable. In a regenerative system, less sorbent is required, less waste produced, and the byproduct sulfur may be used rather than discarded.

Several regeneration processes--atmospheric and high pressure--have been considered (ref. 1). The following pressurized regenerative process reactions have been studied:

| REACTION | APPLICABLE PROCESSES |
|---|---|
| $CaSO_4 + \underset{(4\ H_2)}{4\ CO} \xrightarrow[10\ ATM]{1600^\circ F} CaS + \underset{(4\ H_2O)}{4\ CO_2}$ | Fluidized Bed Combustion |
| $CaS + H_2O + CO_2 \xrightarrow[P \geq 10\ ATM]{T \geq 1300^\circ F} CaCO_3 + H_2S$ | Fluidized Bed Combustion<br>Coal Gasification<br>Oil Gasification |

Results of these tests have been resported (refs. 1,6).

The first step, reduction of calcium sulfate to calcium sulfide, proceeds readily to completion (refs. 6,20) and it is the second step (conversion of calcium sulfide to calcium carbonate) which is crucial to success of the process. TG studies at 10 atmospheres pressure show that calcium sulfide is only partially regenerable and that the nonregenerable fraction increases as the sorbent is cycled between sulfidation and regeneration. The activity of the sorbent in sulfur removal decreases by approximately 3 percent of the sulfidation rate per cycle as shown in figure 7, and is

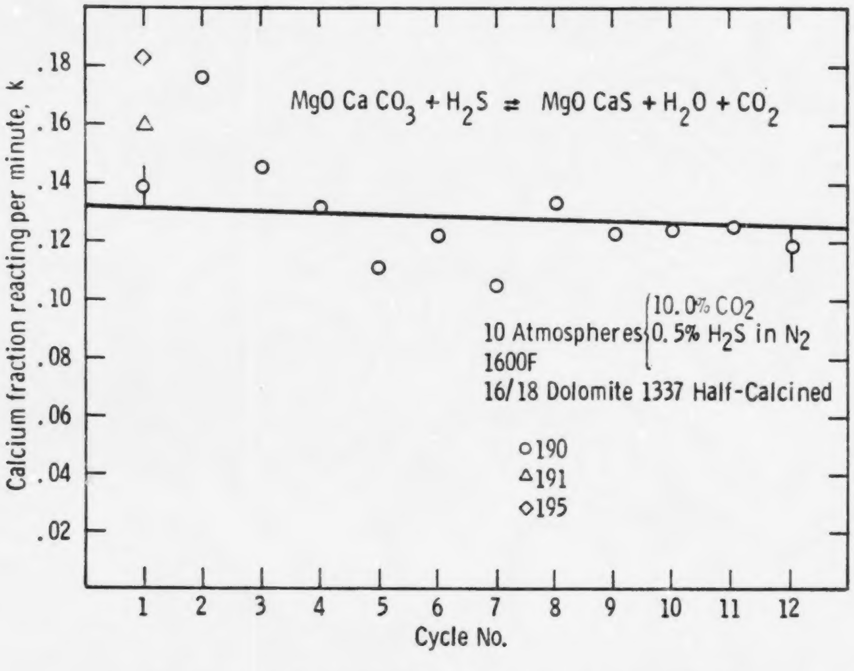

Figure 7 —Rate of sulfidation in cyclic experiment TG 190

apparently not a critical factor. Thus the critical phenomenon in regeneration has been identified as the extent of reaction to form $CaCO_3$ under conditions which yield a sufficient concentration of hydrogen sulfide for recovery in a Claus system. The decrease in regenerability on cycling the sorbent is shown in figure 8, over a 2l-cycle run. Since the stone makeup rate will be partially governed by attrition losses, 20 cycles is considered an acceptable sorbent lifetime. Each mole of calcium transferred 4.7 moles of sulfur to the regenerator during the run and, since it would be discarded from the sorbent cycle in the sulfided condition, can be projected to trap $> 5.5$ moles of sulfur. Decreasing the regeneration temperature would improve the recovered concentration of $H_2S$, but would decrease the regenerability of the stone. It has been noted in the TG experiments that when regeneration rates become negligible, the reaction is revived by increasing the temperature. Further work on this aspect of the problem continues to explore methods for improving the yield of calcium carbonate in regeneration.

In assessing the value of kinetic results obtained by the TG on these processes, it is pertinent to recall that we are determining the conditions under which thermodynamically favorable chemical reactions become rate limiting in the overall process. The projection that good sulfur retention and high utilization of the CaO should be attainable in fluidized beds is simply a statement that the chemical reactions will not of themselves prevent the desired conversion. In the actual situation of a fluidized bed, some other process may intervene to become rate-limiting, such as attrition of the sorbent, coal-ash-sorbent agglomeration, tar deposition on the sorbent, or eutectic formation. However, the probability of overcoming such problems is enhanced when the reaction rate itself is known to be favorable, and can be eliminated from consideration as a source of poor sulfur retention.

## CONCLUSIONS

Low-Btu gasification and fluidized bed combustion can employ high temperature sulfur removal with limestones and/or dolomites to reduce the cost of electrical energy, increase overall plant efficiency, and avoid the generation of environmental problems. A

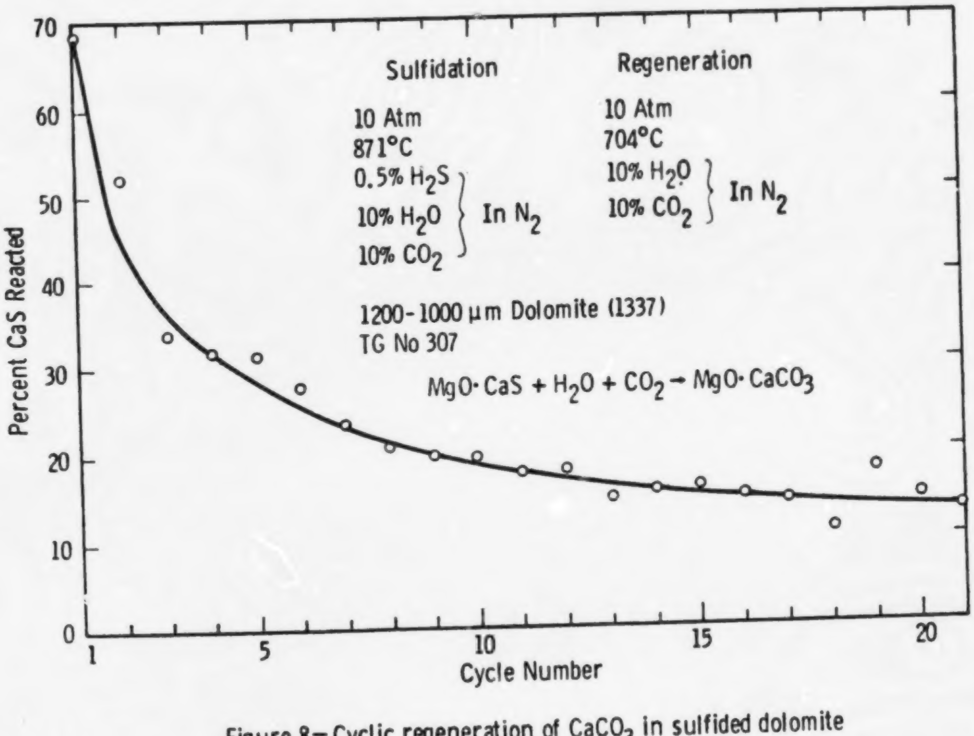

Figure 8—Cyclic regeneration of $CaCO_3$ in sulfided dolomite

thermogravimetric analysis system, modified to operate at high pressures and temperatures, has provided a reliable and efficient method to obtain design data. The results presented show:

(1) High utilization (1.2 to 1.5 Ca/S molar ratio) of calcium in limestones/dolomites for sulfur removal in pressurized fluidized bed combustion can be achieved. This minimizes the sorbent requirement and permits the use of a once-through sorbent process wich has low cost, high efficiency, minimum complexity, and high reliability.

(2) Sulfur removal under reducing conditions approaches the stoichiometric maximum capacity of the sorbent. Sulfur removal under reducing conditions is not a limiting process step.

(3) Regenerability of CaS decreases on cycling (21-cycle test) and represents the limiting process step in the gasification sulfur removal system.

(4) TG systems can be effectively used to investigate fundamental chemical problems, determine the feasibility of processes, assess the effect of system modification, and provide design data.

*Acknowledgments*

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# CLEAN FUELS FROM COAL BY THE COED PROCESS

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*Abstract*

*Present supplies of natural clean fuels are not adequate to meet our growing energy needs. The conversion of coal into synthetic clean fuels would help solve this problem. The COED process has demonstrated its ability to convert practically any type of coal into very-low-sulfur synthetic crude oil, clean fuel gas, and char. The char can be gasified to produce additional clean fuel gas for power generation. Syncrude can be fed to oil refineries or used directly as fuel oil. Essentially all of the sulfur in the coal is converted into hydrogen sulfide which is readily recovered as elemental sulfur by conventional techniques. This paper considers ways in which the COED process can convert coal while satisfying environmental concerns. Processing methods and flows are described for a commercial COED plant to process a high-sulfur, agglomerating coal and a low-sulfur coal.*

## SUMMARY

In a COED plant with added char gasification, over 95 percent of the sulfur in the coal is recovered as elemental sulfur. About 0.5 percent of the sulfur is emitted to the air as $SO_2$, and the remaining sulfur is in the ash. Total recycle of process liquors eliminates water pollution as well as saves water. Ash disposal is a moderate problem, but commercial handling practices are well established.

The COED pilot plant is fully operational. Funding by industry is being sought by FMC for a development program to test the char gasification step in existing processes.

An economic estimate using discounted cash flow indicates that a 25,000 ton-per-day COED plant with added char gasification can produce syncrude at $7 to $9 per barrel and fuel gas at $0.70 to $0.80 per million Btu. This is based on feeding Illinois No. 6 seam-coal at $8 per ton, with an investor's rate of return of 12 percent after taxes (ref. 1).

## INTRODUCTION

Project COED (Char-Oil-Energy Development) was initiated in 1962 when the Office of Coal Research, Department of the Interior, contracted with the FMC Corporation to develop a process for upgrading coal to a synthetic crude oil, a salable gaseous product, and char. Several years of bench-scale research and the operation of a 100 lb/hr process development unit led to the design, construction, and operation of a 36 ton-per-day pilot plant. Figure 1 is a photo of the pilot plant located at the FMC Corporation R&D Center in Princeton, New Jersey. The plant has been in operation since July 1970 and is fully operational. To date, over 18,000 tons of various agglomerating and nonagglomerating coals have been processed through the pilot plant. Several demonstration runs of 30 days in duration were made in the pyrolysis section, while several runs of over 2 weeks in duration were made in the hydrotreating section.

From the data obtained in the 36 ton-per-day pilot plant, a preliminary design of a commercial 25,000 ton-per-day COED plant was made. Environmental aspects of such a plant are studied in this paper for two coals: Illinois No. 6 seam-coal, a Midwestern high-sulfur agglomerating coal; and Utah King coal, a Western low-sulfur bituminous coal. Large reserves of each of these bituminous coal types exist. Typical properties of these coals are shown in table 1.

## PROCESS DESCRIPTION

A schematic of a commercial COED complex is shown in figure 2, and yields from this plant are given in table 2. This flowsheet includes auxiliary plants and shows the various process waste streams. In the COED process, coal is crushed, dried, and then heated to successively higher temperatures in a series of fluidized-bed reactors operated at low pressure (1 to 2 atm.). In each fluidized bed, pyrolysis liberates a fraction of the volatile matter of the coal. The temperature of each bed is just below the temperature at which the coal would agglomerate and plug the bed. Once the coal is partially devolatized in one reactor, it can then be heated further in the next reactor. Typically, four stages operating at 550°, 850°, 1000°, and 1550°F are used. The number of stages and the operating temperatures vary with the agglomerating properties of the coal. Heat for the process is generated by burning char in the fourth stage with a steam-oxygen mixture, and then using hot gases and the hot char from the fourth stage to heat the other vessels.

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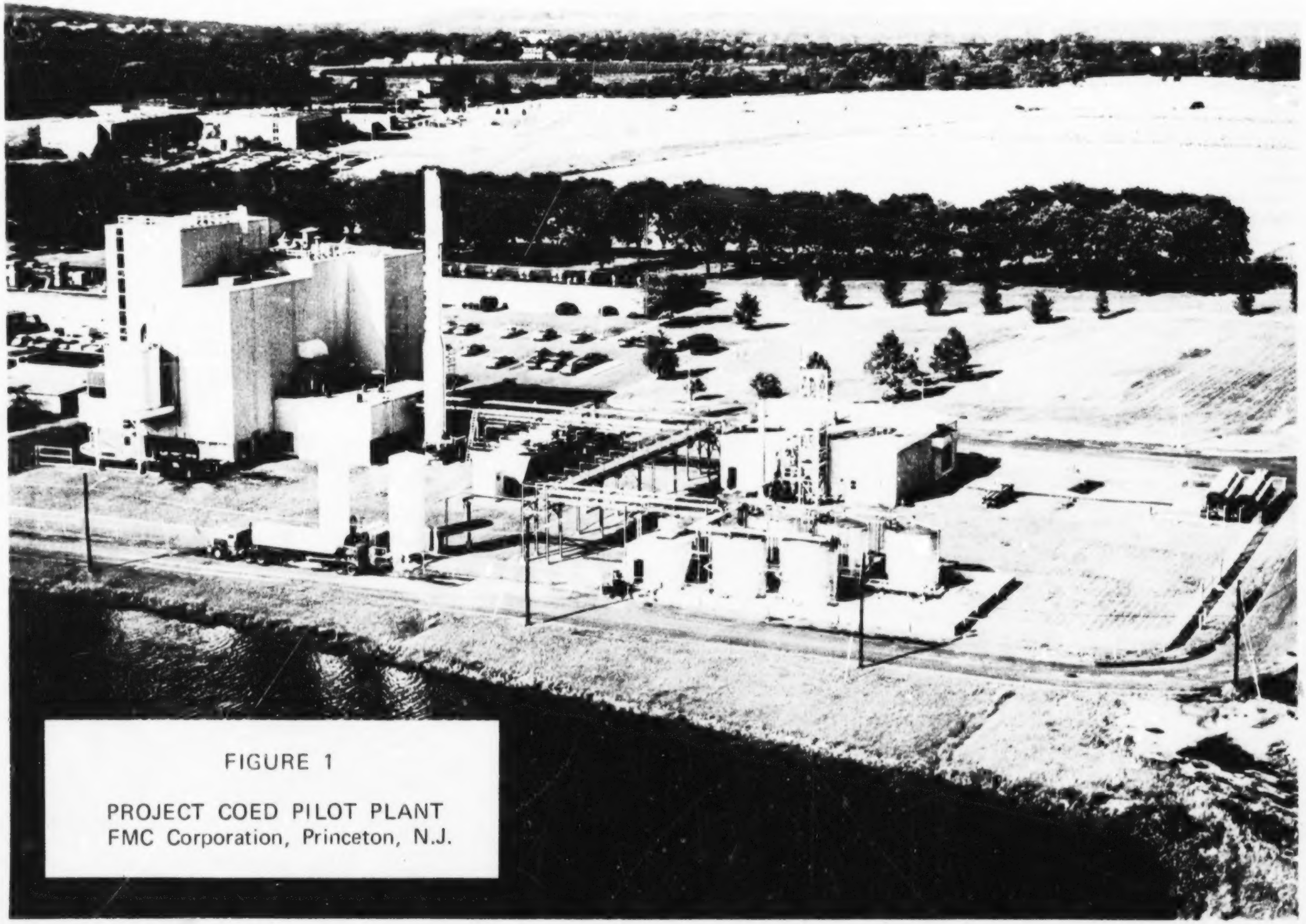


FIGURE 1

PROJECT COED PILOT PLANT
FMC Corporation, Princeton, N.J.

Table 1. Properties of typical coal feeds

| | Utah | Ill. No. 6 |
|---|---|---|
| Bituminous rank | High volatile B bituminous | High volatile B/C bituminous |
| Moisture, wt % | 6.4 | 14 |
| Proximate analysis, wt %, dry | | |
| Volatile matter | 41.7 | 38.1 |
| Fixed carbon | 51.9 | 49.8 |
| Ash | 6.4 | 12.1 |
| Ultimate analysis, wt %, dry | | |
| Carbon | 75.8 | 67.0 |
| Hydrogen | 5.8 | 4.8 |
| Nitrogen | 1.7 | 1.3 |
| Sulfur | 0.6 | 4.1 |
| Oxygen | 9.7 | 10.5 |
| Ash | 6.4 | 12.1 |
| Chlorine | 0.004 | 0.2 |
| Iron* | 0.12 | 1.6 |
| Higher heating value, Btu/lb dry coal | 13,900 | 12,150 |

* Included in "ash" above.

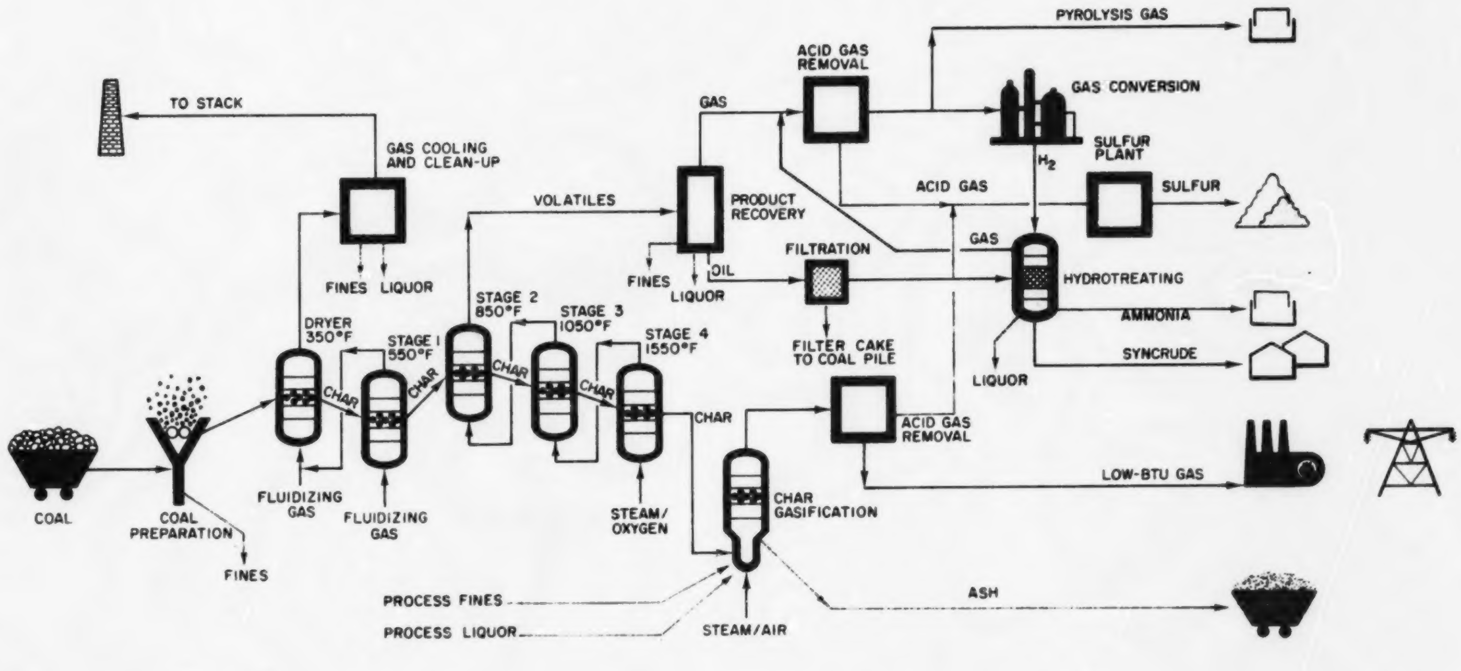
Table 2. Product yields for a commercial COED plant

| | Utah Coal | Illinois No. 6 Coal |
|---|---|---|
| **Feeds** | | |
| Coal, ton/day, dry | 24,000 | 24,000 |
| Oxygen, ton/day | 3,800 | 3,800 |
| **Products** | | |
| Syncrude, bbl/day | 31,700 | 26,000 |
| Net pyrolysis gas, million scf/day[a] | 113 | 112 |
| heating value (HHV), Btu/scf | 510 | 510 |
| Char, ton/day | 13,200 | 12,950 |
| Sulfur, ton/day | 72 | 390 |
| Ammonia, ton/day | 55 | 49 |
| **Products with char gasification added** | | |
| Syncrude, bbl/day | 31,700 | 26,000 |
| Net pyrolysis gas, million scf/day[b] | 202 | 201 |
| heating value (HHV), Btu/scf | 510 | 510 |
| Net low-Btu gas, million scf/day[c] | 1,385 | 1,267 |
| heatng value (HHV), Btu/scf | 170 | 170 |
| Sulfur, ton/day | 159 | 866 |
| Ammonia, ton/day | 55 | 49 |

[a] Gas yield after supplying plant fuel gas and feedstock to the hydrogen plant.
[b] Gas yield after supplying feedstock to the hydrogen plant.
[c] Gas yield after supplying plant fuel gas.

# FIGURE 2

COED PROCESS
WITH ADDED CHAR GASIFICATION
FMC-OCR


TO STACK
GAS COOLING AND CLEAN-UP
FINES
LIQUOR
VOLATILES
GAS
PRODUCT RECOVERY
FINES
OIL
LIQUOR
ACID GAS REMOVAL
PYROLYSIS GAS
GAS CONVERSION
H2
SULFUR PLANT
ACID GAS
SULFUR
FILTRATION
GAS
HYDROTREATING
AMMONIA
SYNCRUDE
LIQUOR
FILTER CAKE TO COAL PILE
COAL
COAL PREPARATION
FINES
DRYER 350°F
CHAR
STAGE I 550°F
CHAR
STAGE 2 850°F
CHAR
STAGE 3 1050°F
CHAR
STAGE 4 1550°F
FLUIDIZING GAS
FLUIDIZING GAS
STEAM/ OXYGEN
CHAR
CHAR GASIFICATION
ACID GAS REMOVAL
LOW-BTU GAS
PROCESS FINES
PROCESS LIQUOR
STEAM/AIR
ASH

The volatile products released from the coal in the fluidized-bed reactors pass to a product recovery system where oil, liquor (water), and gases are separated. The pyrolysis oil is tarry and laden with solids and sulfur. It is filtered to remove solids. The solids-free oil is hydrotreated in a fixed-bed catalytic reactor operating typically at 750°F and 2,000 psig. A nickel-molybdenum catalyst is used. Hydrotreating removes most of the sulur, nitrogen, and oxygen from the oil and cracks it to produce a lighter, more fluid synthetic crude oil. Purge gas and purge liquor streams containing hydrogen sulfide and ammonia are generated in the hydrotreating section. Both streams are cleaned and recycled.

The char product may be burned directly in a boiler if the sulfur content is low enough to meet emissions standards. Since this is not generally the case, one of several existing commercial processes was chosen as the design case to gasify the char. Char is fed at 1,600°F into a fluidized-bed gasifier blown with air and steam. The sulfur comes out in the ash and as $H_2S$ in the gas. Fly ash and $H_2S$ are scrubbed from the gas. Gas streams are cleaned of hydrogen sulfide and carbon dioxide using conventional process such as hot carbonate or Sulfinol. Because the pyrolysis gas is the hydrogen source for hydrotreating, it must be cleaned separately from the low-Btu gas. The hydrotreating bleed gas which is hydrocarbon-rich is blended with the pyrolysis gas for treatment. Part of the cleaned pyrolysis gas is sent to a reforming plant for conversion to hydrogen. A portion of the low-Btu gas is used to fire all furnaces in the plant. The remaining clean pyrolysis gas and clean low-Btu gas are blended and sold as a fuel for a power plant. The hydrogen sulfide is recovered as explained in a subsequent section of this paper.

## PRODUCT USES

### *Synthetic Crude Oil*

COED oil is a full-range synthetic crude oil, capable of being fed to a petroleum refinery and producing a full product slate. Properties of syncrudes from Illinois No. 6 and Utah coals are shown in table 3. An experimental study was carried out by the Atlantic Richfield Company to define those refinery processes where syncrude could best be used. A subsequent study was carried out by Chem Systems, in which their refinery linear programming model showed syncrude to have essentially the same value as sweet domestic crude (ref. 7).

A promising use of the syncrude is to distill it into naphtha and No. 4 fuel oil cuts. The naphtha can be reformed into high-octane gasoline. No. 4 fuel oil is used primarily to fire large boilers, so syncrude was tested in this application. In November 1973, a successful test of powering the Navy's destroyer USS Johnston was made with the COED synthetic crude oil. About 17,000 gallons of syncrude topped to give a fuel with 160°F flash point was burned in a 30-hour shipboard test. Syncrude was also tested as fuel to a small industrial boiler firing 1.5 million Btu/hr. Stack gas emissions were monitored and are reported in table 4. All emissions from syncrude combustion were significantly lower than those from a typical No. 4 residual fuel.

### *Fuel Gas*

Pyrolysis gas is a medium heating-value gas (510 Btu/scf clean gas). Typical analyses of COED fuel gases are shown in table 5. A portion of this gas is used to make hydrogen for hydrotreating. The remainder can be sold as clean fuel gas or can be converted to hydrogen for sale.

Additional low-Btu fuel gas (170 Btu/scf) can be made by gasification of char with air and steam. This would be sold as boiler fuel. The analysis of combined pyrolysis gas and gasifier gas product is given in table 5. Its heating value is 220 Btu/scf.

### *Char*

Properties of chars from Illinois No. 6 and Utah coals are shown in table 6. Chars contain roughly the same concentration of sulfur as the coal, and twice the ash. Most chars must be gasified because they contain too much sulfur for direct combustion.

Operating a COED plant on a low-sulfur coal, such as Utah A seam, produces a char with a sulfur content that is acceptable for combustion in power plants in some States. To demonstrate that COED char would be an acceptable fuel for an industrial boiler, a 3-day test was made in February 1974. An anthracite boiler with a generating capacity of 175,000 lbs/hr of steam was fired with char derived from Utah A seam coal. Operation and turndown of the boiler on the COED char were successfully demonstrated.

## SULFUR BALANCE

Sulfur balances are shown in table 7 for a 25,000 ton-per-day COED plant processing Utah or Illinois coal, with or without char gasification. Nearly half the sulfur in the feed coal comes out in the char for

Table 3. Typical syncrude properties*

| Coal Source | Utah A-seam | Illinois No. 6-seam |
|---|---|---|
| API, °@60°F | 20 | 22 |
| Pour point, °F | 60 | 0 |
| Flash point, PMCC, °F | 75 | 60 |
| Viscosity, cs. @ 100°F | 8 | 5 |
| Ultimate analysis, wt. % | | |
| C | 87.2 | 87.1 |
| H | 11.0 | 10.9 |
| N | 0.2 | 0.3 |
| O | 1.4 | 1.6 |
| S | 0.1 | 0.1 |
| Ash | <0.01 | <0.01 |
| Moisture | 0.1 | 0.1 |
| ASTM distillation | | |
| IBP | 280 | 190 |
| 10% | 430 | 273 |
| 30% | 530 | 390 |
| 50% | 660 | 518 |
| 70% | 780 | 600 |
| 90% | 920 | 684 |
| EP (95%) | 950 | 746 |
| Metals, ppm | <10 | <10 |
| % Carbon residue, 10% bottoms | -- | 4.6 |
| Hydrocarbon type analysis, liquid vol. % | | |
| Paraffins | 23.7 | 10.4 |
| Olefins | 0 | 0 |
| Naphthenes | 42.2 | 41.4 |
| Aromatics | 34.1 | 48.2 |

* Properties depend on severity of operation of hydrotreating unit.

Table 4. No. 4 fuel oil specifications

390-EP COED syncrude
Illinois No. 6 coal

| Test* | Result | Typical No. 4 residual fuel |
|---|---|---|
| Particulates, lbs./100 gal. | 4.6 | 19.2 |
| Gaseous contaminants, ppm | | |
| $SO_x$ | 60 | 490 |
| $NO_x$ | 53 | 95 |
| CO | 4.5 | 6.0 |
| HC | 3.0 | 7.1 |

* Test Conditions: Fuel was burned in a small industrial burner firing a 1.5 million Btu/hr boiler. Stack gas is controlled at 12.5 percent $CO_2$, approximately equivalent to 25 percent excess air and stack temperature is held at 500° to 510°F.

Table 5. Typical analyses of product gases from COED plant with added char gasification

| Gas analysis, mole % $H_2S$, $CO_2$-free gas | Pyrolysis gas | Gasifier gas | Net combined product gas |
|---|---|---|---|
| $N_2$ | 0.6 | 47.1 | 39.6 |
| CO | 21.6 | 36.6 | 34.6 |
| $H_2$ | 55.5 | 15.9 | 22.5 |
| $CH_4$ | 19.3 | 0.4 | 2.7 |
| $C_2H_4$ | 0.5 | -- | 0.10 |
| $C_2H_6$ | 1.4 | — | 0.22 |
| $C_3H_6$ | 0.3 | -- | 0.05 |
| $C_3H_8$ | 0.2 | -- | 0.01 |
| $C_4$+ | 0.6 | -- | 0.11 |
| Higher heating value, Btu/scf | 510 | 170 | 220 |
| Impurities in raw gas, mole % | | | |
| $CO_2$ | 20.9 | 1.6 | -- |
| $H_2S$ (Ill. No. 6 coal) | 1.3 | 0.6 | -- |
| $H_2S$ (Utah coal) | 0.2 | 0.09 | -- |
| $H_2S$ in combined product gas | | | <10 ppm |
| $SO_2$ in stack gas from sulfur plant | | | 200 ppm |

Table 6. Properties of char product

| | Utah | Illinois No. 6 |
|---|---|---|
| Proximate analysis wt %, dry | | |
| Volatile matter | 6.1 | 2.7 |
| Fixed carbon | 80.2 | 77.0 |
| Ash | 13.7 | 20.3 |
| Ultimate analysis wt %, dry | | |
| Carbon | 81.5 | 73.4 |
| Hydrogen | 1.3 | 0.8 |
| Nitrogen | 1.5 | 1.0 |
| Sulfur | 0.5 | 3.4 |
| Oxygen | 1.5 | 1.0 |
| Ash | 13.7 | 20.3 |
| Chlorine | 0.006 | 0.1 |
| Iron* | 0.28 | -- |
| Higher heating value, Btu/lb. dry | 12,310 | 11,040 |

* Included in "ash" above.

Table 7. Sulfur balance
25,000 TPD COED plant with added char gasification

| | Utah coal tons/hr of S | Illinois coal tons/hr of S |
|---|---|---|
| COED section | | |
| Coal (total input) | 6.0 | 41.0 |
| Syncrude | 0.1 | 0.2 |
| Elemental sulfur | 3.1 | 22.4 |
| $SO_2$ emissions* | 0.02 | 0.1 |
| Char (to gasifier) | 2.8 | 18.3 |
| Total output | 6.0 | 41.0 |
| Gasifier section | | |
| Char (total input) | 2.8 | 18.3 |
| Elemental sulfur | 2.6 | 17.8 |
| $SO_2$ emissions* | 0.01 | 0.1 |
| Ash | 0.2 | 0.4 |
| Total output | 2.8 | 18.3 |

* From sulfur-recovery plants.

Table 8. Summary of waste streams and treatment methods

| Waste stream | Sources | Treatment |
|---|---|---|
| Hydrogen sulfide | Raw pyrolysis gas,<br>raw gasifier gas,<br>hydrotreating purge gas | 1. Scrubbing by sulfinol, hot carbonate, etc.<br>2. Claus + tailgas or Stretford |
| Process liquors | Pyrolysis recovery,<br>hydrotreating recovery | 1. Oil skimming<br>2. Clarification (solids removal)<br>3. Recycle to pyrolysis |
| Process condensate (weak liquors) | Coal drying<br>gasifier offgas | 1. Clarification (solids removal)<br>2. Biodegradation and/or recycle as low-quality process steam |
| Ash | Gasification | 1. Dampen<br>2. Return to spent mine site |
| Fines (dust) | Coal preparation<br>char handling<br>pyrolysis liquor clarifiers<br>pyrolysis external cyclones<br>raw-oil filters | Feed dry fines to gasifier or into char product; recycle oily fines to coal feed |
| Sulfur dioxide | Sulfur recovery plant | Treatment not needed—within acceptable limits |

either high- or low-sulfur coal. The char gasifier releases most of this sulfur as hydrogen sulfide, with the rest retained in the ash. Over 99 percent of the $H_2S$ is recovered as elemental sulfur. Hydrotreating removes over 95 percent of the sulfur from Illinois oil and over 80 percent from the low-sulfur Utah oil.

## TREATMENT OF WASTE STREAMS

Treatments of the major waste streams from the COED process are summarized in table 8 and are explained below.

### *Sulfur Scrubbing and Recovery*

Offgases from pyrolysis and hydrotreating contain $H_2S$, which is scrubbed out of the combined gas. Any of several commercial processes can be used satisfactorily to scrub the $H_2S$ and most of the $CO_2$ out of the product gases. Chemical absorption processes such as Sulfinol or hot carbonate are preferred because they can operate at low pressure, thus minimizing compression cost. Acid gas is collected in a solvent, then flashed off and sent to the sulfur recovery plant. All of these processes can leave as little as 5 to 10 ppm $H_2S$ in the product gas.

It is likely that the char will be gasified. This process liberates sulfur from the char as $H_2S$ and possibly as small amounts of COS (carbonyl sulfide). Gasifier offgas passes through an acid-gas scrubbing plant like the one described above. Carbonyl sulfide is produced mainly in high-temperature (slagging) gasifiers. Any of the chemical absorption processes absorb COS, although it degrades amine solvents. The Sulfinol solvent is resistant to COS degradation. Gases containing CO and $H_2$ have been treated successfully by amine systems, so COED gases present no problem when applying these systems (refs. 2,3,4).

Elemental sulfur is recovered from the acid-gas streams using existing commercial processes. High-sulfur coals, such as Illinois No. 6, yield concentrated $H_2S$ streams (10 to 20 percent $H_2S$) which are treated in a Claus plant. The tail gas is further treated by a method such as the Beavon or SCOT* processes to limit $SO_2$ in effluent gases to 200 ppm. Over 99 percent of the sulfur is recovered in this manner. Utah coal yields acid-gas containing 5 percent $H_2S$. This stream is treated by the Stretford process to recover about 99 percent of the sulfur (refs, 2,5,6).

### *Process Liquors*

Process liquors emerge from three areas of a COED plant. Typical analyses of these streams are given in table 9.

A fairly clean condensate liquor comes from the coal-drying and first-stage pyrolysis section. This comes primarily from inherent moisture in the coal feed. Fines are removed in a clarifier, and the liquor is recycled to generate low-pressure steam for the process. A fraction of the liquor is taken as boiler blowdown, and is injected into the char gasifiers. The pH of this stream is 7 to 8.

Pyrolysis liquor comes from the oil recovery section of the pyrolysis plant. It contains tars, fines, phenolics, ammonia, and hydrogen sulfide. Tars are removed in an oil separation tank, and fines are removed in a clarifier. This stream can contain as much as 1.0 percent dissolved organics. The preferred treatment is to recycle the material and pyrolyse it. This is done by partially flashing the liquor and injecting it as liquid and vapor into either the gasifiers or the hottest pyrolyser. Total liquor recycle is possible because more water is consumed in the hottest pyrolysis vessel than is produced in the intermediate stages. A biological degradation pond is provided to handle liquors from process upsets.

Oil hydrotreating produces water, ammonia, and hydrogen sulfide byproducts, which leave the plant primarily in a liquor stream. This liquor is supersaturated with $H_2S$ and $NH_3$ as it leaves the

Table 9. Properties of process liquors

| Coal | Utah | Illinois No. 6 |
|---|---|---|
| First stage pyrolysis liquor (weight percent) | | |
| Carbon | 0.17 | -- |
| Nitrogen | 0.01 | 0.05 |
| Sulfur | 0.01 | 0.07 |
| Phenol | 9.2 ppm | 0.00 |
| Entrained oil | 0.01 | -- |
| Suspended solids | 0.03 | 0.49 |
| pH | 8.1 | 3.6 |
| Second stage pyrolysis liquor | | |
| Carbon | 2.0 | -- |
| Nitrogen | 0.88 | 0.93 |
| Sulfur | 0.13 | 0.18 |
| Phenol | 0.40 | 0.38 |
| Entrained oil | 0.2 | 0.0–0.5 |
| Suspended solids | 0.64 | 1.09 |
| pH | 9.0 | 8.8 |
| Hydrotreating liquor | | |
| Carbon | 1.7 | 0.8 |
| Nitrogen | 5.2 | 5.0 |
| Sulfur | 4.2 | 8.7 |
| pH | 11.5 | 9.3 |

---

*Shell Claus Offgas Treating.

## TABLE 10

## Project COED Technical Reports Available from the U.S. Government

"Char Oil Energy Development" - Project COED, Final Report
Period Covered: June 1962 - December 1965
R&D Report No. 11
OCR Contract No. 14-01-0001-235
Refer to: PB-169562 (Volume I) - $6.00*
PB-169563 (Volume II) - $6.00*

"Char Oil Energy Development" - Project COED, Amendment No. 3,
Final Report, Period Covered: January - October 1966
R&D Report No. 11
OCR Contract No. 14-01-0001-235
Refer to: PB-173916 (Final) $6.00*
PB-173917 (Appendix) - $6.00*

"Char Oil Energy Development" - Project COED, Interim Report No. 1,
Period Covered: September 1966 - February 1970
R&D Report No. 56
OCR Contract No. 14-01-0001-498
Refer to: Titled report and
GPO Catalog No. 163.10:56/int 1
Cost: $2.50**

"Char Oil Energy Development" - The Desulfurization of COED Char,
Part III" - Project COED, Interim Report No. 2,
Period Covered: December 1968 - May 1970
R&D Report No. 56
OCR Contract No. 14-01-0001-498
Refer to: Titled report and
GPO Catalog No. 163.10:56/int 2
Cost: $1.25**

"Char Oil Energy Development" - Project COED, Final Report,
Period Covered: October 1966 - June 1971
R&D Report No. 56
OCR Contract No. 14-01-0001-498
Refer to: Titled report and
GPO Catalog No. 163.10:56
Cost: $4.00**

"Char Oil Energy Development" - Project COED
Period Covered: July 1971 - June 1972
R&D Report No. 73 - Interim Report No. 1
OCR Contract No. 14-32-0001-1212
Refer to: Titled report and
GPO Catalog No. 163.10:73/int 1

"Char Oil Energy Development" - Project COED
Period Covered: July 1972 - June 1973
R&D Report No. 73 - Interim Report No. 2
OCR Contract No. 14-32-0001-1212
Refer to: Titled report and
GPO Catalog No. (not available at the present time)
Cost: Not established

Instructions for ordering reports. All reports must be prepaid.

*Available from:

U.S. Department of Commerce
National Technical Information Service
5285 Port Royal Road
Springfield, Virginia 22151
Attention: Storage and Dissemination Section

(Make check payable to: National Technical Information Service)

**Available from:

Superintendent of Documents
U.S. Government Printing Office
North Capitol and H Streets, N.W.
Washington, D.C. 20401

(Make check payable to: Superintendent of Documents) pressure vessels, so it is passed through a stripping tower in which product gases pick up most of the $H_2S$ and $NH_3$. This gas is recycled to the acid-gas removal plant. The stripped liquor contains dissolved organics at a level similar to pyrolysis liquor. Therefore, the hydrotreating liquor is blended with pyrolysis liquor and recycled, as previously explained.

A COED plant without char gasifiers needs biological liquor-treatment facilities. The pyrolysis and hydrotreating liquors can still be pyrolysed in the hottest pyrolyser. The dryingsystem liquor stream is too large to be added to this pyrolyser, so it is biologically treated. The organic content of this stream is low, and treatment is not difficult.

### *Ash*

Typically, about 10 percent of the coal feed is ash which must be disposed of as it emerges from the gasifiers. Granular ash and flyash streams are combined and moistened to stop dusting, and are shipped to the mine as landfill for spent minesites. The finest flyash is collected by wet scrubbing. The wet concentrate is used for ash moistening.

A COED plant without char gasifiers puts out all the ash in the char product. The power plant handles the ash in this case.

### *Fines*

Dust problems are controlled with cyclones and bag filters in the coal preparation and char handling facilities. Fines from the pyrolysis plant are collected by cyclones, both internal and external to the vessels. Fines escaping from cyclones are collected by wet scrubbers and concentrated in a clarifier. Both wet and dry fines are fed into the gasifiers. Raw-oil filtration yields a filter cake containing oily fines. This material is returned to the coal pile for recycle.

### *Process Cooling*

All process cooling is done using wet cooling towers. The total plant water requirement is about 15,000 acre-feet per year for a plant with added char gasification. Roughly 10,000 acre-feet per year are used for cooling. Since low-quality process water is needed, no water effluents are given off, either from cooling or from the process itself. Recycle of process water saves about 9 percent of the total water requirement.

## ACKNOWLEDGMENT

The authors acknowledge the guidance and support from the Office of Coal Research. Their contributions of technical support and funding have helped to make the COED pilot plant a success. A list of technical reports available from the U.S. Government on Project COED is attached in table 10.

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# ENVIRONMENTAL ASPECTS OF THE SRC PROCESS

C. R. Hinderliter*

*Abstract*

*The Solvent Refined Coal (SRC) Process is being developed by the Pittsburg & Midway Coal Mining Co. under the sponsorship of the Office of Coal Research. In the SRC process, coal is first dissolved under moderate hydrogen pressure in a heavy aromatic solvent. The resultant coal solution is filtered to remove ash and a small amount of insoluble organic material and is fractionated to recover the solvent. The product is a heavy residual fuel that is solid at ambient conditions and that contains less than 0.8 percent sulfur and less than 0.1 percent ash. The yield of SRC from coal is approximately 60 percent by weight; in addition, about 15 percent of the coal is converted into light liquids and gases.*

*An SRC plant would have to deal with several potential sources of pollution. Coal must be handled and pulverized in a manner designed to minimize emission of coal dust. The sulfur removed from the coal by processing is in the form of hydrogen sulfide which must be absorbed from product gases and converted to elemental sulfur. The sulfur plant itself must include tail gas cleanup facilities to reduce sulfur emissions. Certain fractions of the light liquids produced will contain substantial amount of phenolics and amines. Process water will be contaminated with these materials and will have to be treated to prevent water pollution. Problems similar to these are faced by other industries and are handled adequately with proven technology. I am confident that with power design an SRC plant can be built that will meet environmental standards.*

## Introduction

This Nation's need to develop new processes to produce clean fuels from coal has been discussed by many people. While developing these new processes, we must insure that the processes themselves are clean and do not have an adverse effect on the environment. This paper discusses one such process called the Solvent Refined Coal Process which is being developed by The Pittsburg & Midway Coal Mining Company (P&M) under the sponsorship of the Office of Coal Research (OCR). This process has been under development since 1962, and the construction of a 50-ton-per-day pilot plant at Fort Lewis, Washington, was recently completed. Although definite commercialization plans have not been finalized for this process, both P&M and OCR are studying the environmental aspects of the Solvent Refined Coal Process. Our discussion today will principally dwell on our concept of a commercial plant which differs substantially in design from the pilot plant. The pilot plant does include many safeguards to protect the environment.

## Background

In 1962, the Office of Coal Research awarded a research contract to Spencer Chemical Company to study the technical feasibility of a coal deashing process. This work was concluded in 1965 with the successful demonstration of the process in a 100-pound-per-hour continuous flow unit. During the term of this contract, Gulf Oil Corporation acquired Spencer Chemical Company and, upon reorganization, reassigned the project to the Research Department of P&M. This deashing process is now called the Solvent Refined Coal (SRC) Process. In 1966, a contract was awarded to P&M to further develop the SRC process through design, construction, and operation of a pilot plant to process 50 tons of coal per day. The design for the pilot plant was completed by Stearns-Roger Corporation in 1969 but funds were not immediately available to plan for construction. In late 1971, sufficient funds became available to plan for construction and Rust Engineering Company was selected, through competitive bidding, to complete the detailed engineering and to construct the pilot plant at Fort Lewis, Washington. In June 1972, OCR extended the contract with P&M to meet the projected needs for construction and operation of the pilot plant. Construction was started in July 1972 and the total cost of the pilot plant has been estimated to be about $20 million.

The pilot plant construction progress has been somewhat slower than scheduled. Completion was initially scheduled for November 1973, but the necessary addition of biological waste water treatment facilities and problems encountered in fabricating the dissolvers delayed completion until April 1974. The biological treatment facilities will still not be completed for several months. The pilot plant will startup using a petroleum-derived solvent, a cycle oil from an FCC unit, and bituminous coal from Western Kentucky. Our laboratory had previously worked almost exclusively with anthracene oil as a solvent, but due to the limited availability of this material, the pilot plant will use a petroleum-derived aromatic oil as startup solvent. To date, work in the laboratory indicates that petroleum-derived solvents are not as well suited as anthracene oil for solution of coal, and different reaction conditions must be used during startup. Most of the recent laboratory effort has been directed to defining reaction conditions for the pilot plant during the startup period. Plans are being made to obtain different coals for testing and for making combustion tests on the SRC product.

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### Process Description

A schematic flow diagram of the SRC process is shown in figure 1. In the SRC process, coal is pulverized and mixed with a solvent to form a slurry. This slurry contains between 25 and 35 weight percent coal. The slurry is pressurized to approximately 1,000 psig, mixed with hydrogen, and heated to approximately 425°C where the coal solution reactions are completed in about 20 minutes, during which time nearly all of the organic matter in the coal dissolves in the solvent. The reaction product is flashed to separate gases, and the liquid is filtered to remove the mineral residue which consists of ash and undissolved coal. The liquid is then fractionated to recover the solvent which is recycled to slurry more feed coal. During each pass through the reactor, a small amount of solvent is converted to light liquids and gases; however, this loss is more than balanced by the amount of coal which is converted to solvent with the net result that the process does not require solvent feed except during initial startup. The liquid remaining after the solvent has been recovered is a heavy residual fuel called "solvent refined coal" that contains less than 0.8 percent sulfur and less than 0.1 percent ash and that solidifies upon cooling to ambient conditions. The solvent refined coal has a melting point of about 350°F and has a heating value of about 16,000 Btu per pound regardless of the quality of the coal feedstock. Smaller quantities of hydrocarbon gases and light distillate liquids are also produced. The solvent refining process removes all of the inorganic sulfur and 60 to 70 percent of the organic sulfur in the coal.

In the present concept of the SRC process, the mineral residue which may contain more than 6 percent sulfur would be recovered from the filters and gasified along with additional coal to make the hydrogen needed for the process. An excess of hydrogen would be made which along with the hydrocarbon gases produced would supply the fuel needed by the process. The sulfur that is removed from the coal appears as hydrogen sulfide in the SRC product gases and the gasifier product gas. The hydrogen sulfide is separated by amine absorption or a similar process and converted to elemental sulfur. The ash in the original coal would be processed eventually through the gasifier and, assuming that a slagging gasifier would be used, this ash would be converted to a clean slag.

### Environmental Considerations

Carnegie Mellon University recently completed a study for P&M in which they compiled Federal and State environmental regulations that may apply to an SRC plant located in States bordering the Ohio River. This report has been submitted to the Office of Coal Research for publication as R&D Report No. 53, Interim Report No. 5. An SRC plant would have to meet both Federal and State air quality standards. There are two types of Federal air regulations: ambient air quality standards and new source performance standards. Ambient air quality standards define the minimum allowable quality of the air in regard to six different pollutants. These standards are given in table 1. New source performance standards have not been enacted for coal refining plants. In the absence of Federal new source performance standards, an SRC plant would have to conform to the State or local regulations that are applicable.

The Federal Water Pollution Control Act amendment of 1972 sets forth as a national goal the complete elimination of all discharges of pollutants by 1985 with particular emphasis on control of toxic materials. New source water quality standards have been proposed for petroleum refineries and for coke plants but not for coal refineries. These proposed Federal standards are very stringent, but certain State regulations are even more limiting. For example, Pennsylvania regulations forbid the discharge of any phenol. It is not certain that existing technology can meet the proposed water quality standards; however, coal refining should have no unique problem and no

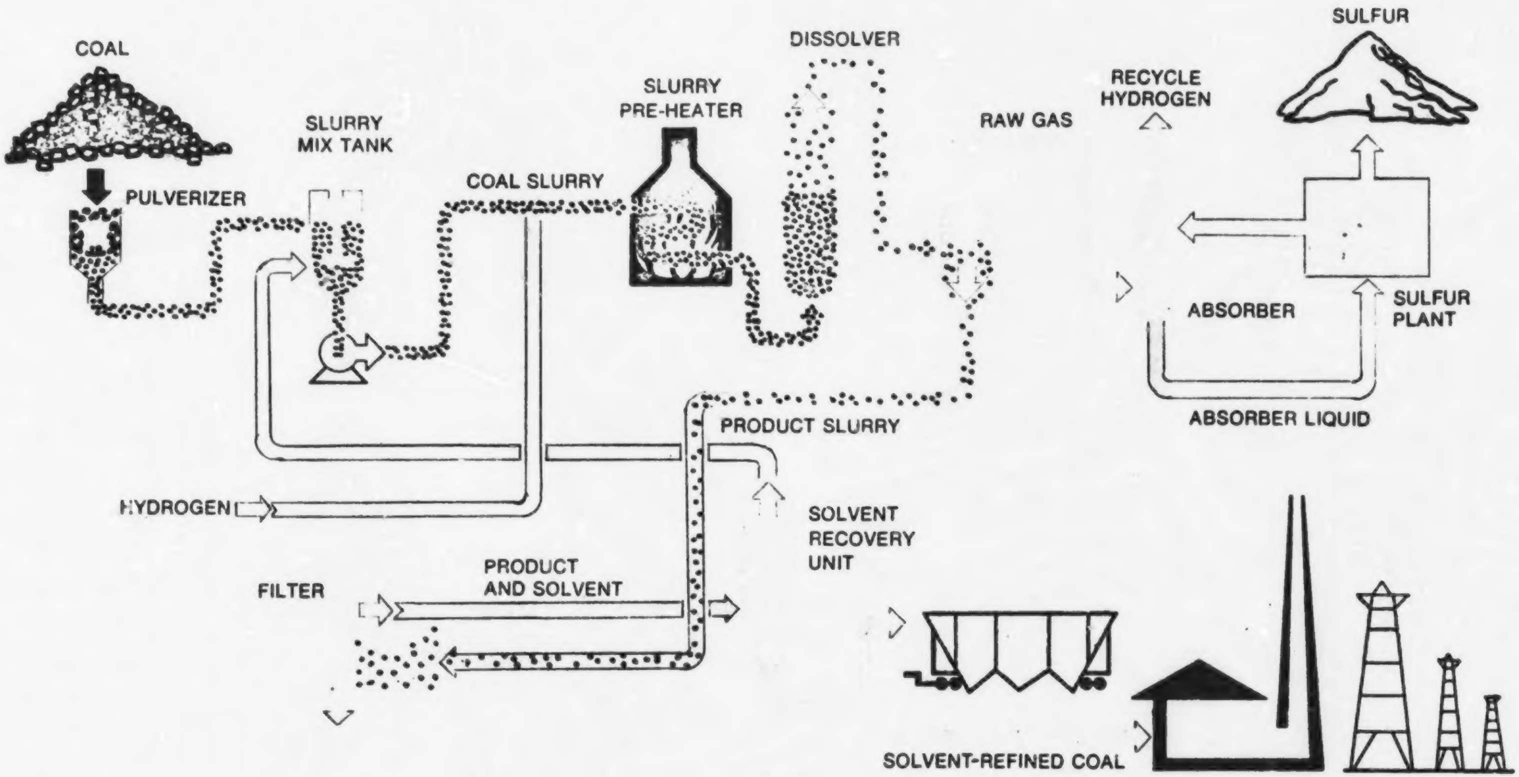
COAL
PULVERIZER
SLURRY MIX TANK
COAL SLURRY
SLURRY PRE-HEATER
DISSOLVER
RAW GAS
RECYCLE HYDROGEN
SULFUR
ABSORBER
SULFUR PLANT
ABSORBER LIQUID
PRODUCT SLURRY
HYDROGEN
SOLVENT RECOVERY UNIT
FILTER
PRODUCT AND SOLVENT
SOLVENT-REFINED COAL

Figure 1. SOLVENT-REFINED COAL PROCESS

Table 1. National Ambient Air Quality Standards

| Contaminant | Averaging interval | Primary standard µg/m³ | Primary standard ppm (by vol.) | Secondary standard µg/m³ | Secondary standard ppm (by vol.) |
|---|---|---|---|---|---|
| Suspended particulates | 1 year | 75 | -- | 60 | -- |
| | 24 hr | 260 | -- | 150 | -- |
| Sulfur dioxide | 1 year | 80 | 0.03 | -- | -- |
| | 24 hr | 365 | 0.14 | -- | -- |
| | 3 hr | -- | -- | 1,300 | 0.5 |
| Carbon monoxide | 8 hr | 10,000 | 9.0 | 10,000 | 9.0 |
| | 1 hr | 40,000 | 35.0 | 40,000 | 35.0 |
| Photochemical oxidant | 1 hr | 160 | 0.08 | 160 | 0.08 |
| Hydrocarbons (nonmethane) | 3 hr (6-9 a.m.) | 160 | 0.24 | 160 | 0.24 |
| Nitrogen dioxide | 1 year | 100 | 0.05 | 100 | 0.05 |

Source: Report by Carnegie Mellon University.

greater difficulty in meeting the water quality standards than would a petroleum refinery and even less difficulty meeeting environmental standards than would a coke plant. Environmental considerations in the design of an SRC plant would primarily focus on air and water pollution, but they are not considered in this paper. In view of the developmental state of the SRC process, a precise definition cannot be given for many areas of such a plant, for this reason, environmental problems are discussed in general terms.

*Coal Preparation*. Coal preparation includes transporting the coal from storage, then pulverizing, drying, and mixing the coal with solvent to form a slurry. There are two potential sources of pollution in these operations. The most obvious is fugitive particulate matter which will result from handling the dry coal. The coal dust problem can be controlled by using covered conveyors and induced draft vents with the vent gas being filtered to remove particulates. If the coal must be dried, the off-gas from the drier also must be filtered to remove particulates. Some water will be liberated from the coal during pulverizing and during slurry preparation; if this water is condensed, it may have to be treated to remove suspended solids and to adjust acidity before discharge. We would not anticipate any difficulty in designing the coal preparation area for an SRC plant since coal preparation is a well-known operation, and we should encounter no surprises. Pilot plant operation should define the slurry preparation operation and determine the extent of coal drying required for optimum operations.

*Coal Solution*. Most of the actual processing will be conducted in an enclosed, pressurized system with equipment failure or leakage being the only potential source of emissions. The coal slurry is pressurized, mixed with hydrogen, and heated to reaction temperature where it is retained for a certain amount of reaction time. After the coal slurry has undergone reaction, the solution is reduced somewhat in temperature and pressure, and the gas is separated from the liquid slurry product. The gas is then cooled to condense water and light liquids. This product gas contains hydrogen sulfide which represents the sulfur removed from the coal and which must be recovered to prevent sulfur emission to the atmosphere. Technology already exists that is adequate to recover this hydrogen sulfide. e.g., amine and carbonate scrubbing processes. This product gas contains a substantial amount of unreacted hydrogen, and a part of the gas may be recycled through the coal solution processing step. The gas which is not recycled would be used for process fuel, but part of it could be reformed to convert the contained hydrocarbon gases to hydrogen for use in the process. An excess of gas would always be made, and this excess gas would be burned to provide heat for the process.

The water that is condensed from the product gas presents a somewhat more difficult problem, one that we have not completely defined. This water will be in contact with the light liquids which contain phenols and amines, some of which will dissolve in the water. Due to the presence of phenols, it would not be possible to directly discharge this water unless it received extensive treatment such as biological processing. An alternate approach to the disposal of this contaminated water would be to use it in the production of hydrogen either by gasification of the mineral residue or coal, or by reforming the product gases. We feel that this alternate is feasible and would be the preferred approach. It may be necessary to remove part of the phenolics from the water before reuse; however, phenolics potentially represent a valuable byproduct that may be recovered for sale in any case. The water requirements for hydrogen manufacture will be greater than the amount of water made by the process, and fresh water makeup would be required to supplement the process water.

*Mineral Residue Recovery and Hydrogen Manufacture*. After separation of gases, the liquid slurry product is filtered to remove the ash and undissolved coal. This solid is referred to as the mineral residue, and it generally will be about one-half carbon and one-half ash and may contain more than 6 percent sulfur. This mineral residue will be removed from the filter; it may be dried and then gasified to make hydrogen for the process. If drying is needed, the dryer must be designed to prevent fugitive emissions which would pollute the air.

The gasifier would convert the mineral residue, which is high in sulfur, into a relatively clean slag and synthesis gas. A slagging gasifier similar in concept to a Bituminous Coal Research Bi-Gas gasifier would be preferred for this operation, although any commercial gasifier could be used. The sulfur in the mineral residue would be converted into hydrogen sulfide which would appear in the synthesis gas and which would be removed by amine or carbonate scrubbing.

It would also be necessary to gasify some coal to make hydrogen, since gasification of the mineral residue would not produce enough hydrogen for the process. An alternate scheme—which could be used if a reliable gasifier of proven design were not available or if the cost of gasification were excessive— would be to re-form part of the product gas to make hydrogen. In this case, an alternate use must be found for the mineral residue, or it would have to be discarded. This scheme would result in a deficiency in fuel gas for plant fuel, and part of the liquid or solid product would be used for plant fuel.

*Sulfur Plant*. The sulfur liberated from the coal by processing appears as hydrogen sulfide in various gas streams. The hydrogen sulfide would be recovered by an acid gas absorption unit and converted to elemental sulfur. A Claus sulfur plant would probably be used with an appropriate tail gas cleanup unit to increase the recovery of sulfur to 99 or 99.5 percent and reduce sulfur emissions to acceptable levels as dictated by environmental regulations. The spent gas from the tail gas unit would be incinerated to insure that any small amount of unrecovered hydrogen sulfide is converted to sulfur dioxide prior to venting to the atmosphere. Proven technology exists with which to build a workable sulfur recovery plant, and no surprises would be expected.

*Process Heaters*. An SRC plant will have several fired heaters which will require substantial quantities of fuel. The slurry preheater and the solvent recovery areas will be the major fuel consumers. The SRC process makes some byproduct gases which would be recovered and used as fuel for the process heaters, as discussed earlier. The gases made would be diluted with unreacted hydrogen and would have a heating value of about 300 to 400 Btu/SCF. The crude gas would contain hydrogen sulfide which would have to be removed by an acid gas absorption unit to a low concentration, such that the gas could be burned without exceeding sulfur oxides emission standards. The fired heaters would also emit some oxides of nitrogen as byproducts of combustion. The fuel gas would contain essentially no combined nitrogen, and the $NO_x$ emissions would depend upon the burner characteristics. It should not be a difficult task to design the fired heaters to be in compliance with $NO_x$ emission standards. In certain process areas, spillage or equipment leakage could result in accumulation of phenolics and other organic materials that could contaminate rain water runoff. It would be desirable to separately collect water runoff from these certain areas of the plant and to use appropriate treatment to guard against discharge of phenolic pollutants from the plant. Since phenolic materials would be present which could contaminate the waste water from an SRC plant, biological waste water treatment facilities would be needed to destroy phenols and to prevent their discharge from the plant. Additional waste treatment facilities would be needed to remove solids and to neutralize the water prior to discharge.

An SRC plant would require a thermal oxidizer to insure incineration of any gaseous discharge from the process. Some process waste streams will contain minor amounts of hydrogen sulfide or carbon monoxide that could not be discharged into the atmosphere without incineration. Certain liquid waste streams, mainly contaminated water, would also be incinerated to destroy organic compounds and to prevent water contamination.

In summation, an SRC plant will have few environmental problems that cannot be handled adequately with existing off-the-shelf technology. The problems that will exist may be presented in a somewhat unconventional manner, but I am confident that environmental standards can be met with foresight and proper design.

# ENVIRONMENTAL ASPECTS OF SOLVENT REFINING

**W. B. Harrison***

*Abstract*

*An effort will be made to analyze the environmental aspects of solvent refining as a strategy to enable the electric utility industry to utilize high sulfur bituminous coal for power generation. Comparisons will be made with other strategies for using such coal, including flue-gas processing and gasification. A brief assessment of the environmental problems of a solvent refining plant will be made.*

In view of the fact that the preceding speaker has addressed some of the specific environmental aspects of a representative solvent refining plant, I wish to discuss environmental aspects from a different point of view. I shall try to place in perspective the kinds of questions which an electric utility company would address in outlining an environmental impact statement for a plant that is to be fueled with solvent refined coal. In effect, the exercise will involve comparing broad issues of environmental interaction of alternatives for meeting the energy demand with some reference to the environmental aspects of each alternative. I presume that there is no need at this conference to develop a statement on the importance of coal in the context of our future energy requirements. From the electric utility viewpoint, it would appear that coal must remain as a major energy resource throughout the remainder of this century, and beyond.

The constraints imposed on the use of coal are characterized as being legal, logistic, technical, and economic. Legal constraints are mainly in the form of State and Federal laws and regulations. In this discussion, reference is made particularly to the environmental regulations which apply both to existing and to new power plants and which govern both ambient air quality and standards of performance (emission limitations). Logistic constraints are best understood by considering the location and character of coal reserves throughout the nation with respect to the location and size of generating plants needing the fuels, the length of supply lines, and the available transportation modes. A related topic, logistic in nature, is the ease with which fuels may be brought into processing plants or with which byproducts and other wastes from the plants may be further treated or stored. The technical constraints are used merely to recognize that any viable option must be based on reliable and reasonable technology. And finally, economic constraints must provide an umbrella over the whole mix of options because some things which are technically or logistically feasible may not be economically feasible.

It is apparent from an examination of the legal constraints that many existing plants and all future plants will require major deviations from practices of the past. The sulfur dioxide regulations provide a specific example of how regulations will influence the operating options faced by a generating plant. Clearly, one option is to select a fuel containing such small amounts of sulfur that the sulfur dioxide regulations can be met. It is apparent that low-sulfur coal is not available in sufficient supply to make this alternative possible in some locations. Therefore, the options are reduced to processing high sulfur fuel in order to remove sulfur before combustion, to processing the flue gas in order to remove sulfur dioxide after combustion, or perhaps to using some combination of the two processes. Sulfur abatement measures that might take place during combustion such as in proposed fluidized-bed combustion boilers are regarded at this point as unproven technology and are, therefore, not considered as viable alternatives. The other options being considered are not necessarily judged to be proven, but at least they are further developed than fluidized-bed technology, and comparisons may be made on the assumption that reliable technology in fuel processing or flue gas processing will emerge in the course of a few years.

In order to illustrate the point of this discussion, it is assumed that two fuel processing options will be available for utility applications: low-Btu gas or solvent-refined coal. The availability of flue gas processing is also assumed, but the particular process is entirely arbitrary for purposes of this discussion. As we move forward in developing these thoughts, the next step will be to identify certain of the factors that would enter into an overall environmental appraisal of the three options—low-Btu gas, solvent-refined coal, or stack gas processing—in the context of achieving adequate sulfur removal from a typical Western Kentucky or Southern Illinois coal for a power plant application.

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Presumably, the total amounts of sulfur to be removed from either the fuel or the stack gas would be comparable irrespective of the strategy selected. The major differences in comparing the three alternatives will lie in the chemical or physical form in which that sulfur may be found. For example, it would appear that the sulfur removed during either gasification or solvent refining may leave the process either as concentrated sulfuric acid or elemental sulfur. The choices are dependent on the potential markets that might exist for either form, but neither form is dictated by the features of the fuel processing. On the other hand, in the stack gas processing alternative, not only is the chemical form of the sulfur dictated by the nature of the process, but also the disposal of the sulfur becomes a problem at the power plant site. Thus, the nature and chemical form of the sulfur arising from each of these alternatives has its own specific environmental impact, and this could influence the feasibility of either option for a specific generating site. The question of location with respect to the generating site perhaps deserves more elaboration. Clearly from the point of view of the power plant operator, either alternative for making clean fuels from coal has advantages over stack gas cleaning because the environmental issues pertaining to fuel processing would likely be the concern of someone else, but the environmental issues of stack gas cleaning have to reside with the power plant operator. This also has a relationship to the question of low-Btu gasification which, because of the low energy density of the product gas, must be accomplished adjacent to or very close to the power plant site. Thus, solvent refining has the greatest versatility with respect to the questions of producing the sulfur in a stable, innocuous form, and the freedom of locating the processing plant at whatever location appears to be most feasible with respect to the source of coal, the existing or most attractive future transportation network, the markets for byproducts, and so forth. Another aspect is that transportation savings are in proportion to the nearness of the processing plant to the mine. This means that solvent refining could offer large transportation savings not applicable to gasification. It also follows that flue gas processing would create additional transportation expenses in comparison, since large quantities of processing materials would have to be shipped in to the power plant site.

In the context of retrofitting an existing plant in order to meet some particular sulfur dioxide abatement goal, there are many plants in which available space limits the choice. In a sense, this is an environmental question. I can visualize many existing power plant sites which simply would not have sufficient room for the installation of flue gas cleaning processes or which would not have space available adjacent to the plant site for the construction of gasification facilities. In such situations, solvent refined coal offers an apparent solution, because retrofitting could be accommodated with very small modifications to the plant, and existing storage and handling facilities could be utilized. Solvent refining offers an alternative to premature obsolescence for such plants.

Still another factor for consideration has to do with the interaction of the alternative on the operations of the power plant itself. For example, flue gas processing requires of the power plant operating staff some skills and disciplines which are not customarily found at such facilities. This would necessarily enlarge the scope of operator training and the breadth of skills required in plant operations and control. Flue gas processing brings also to the plant large quantities of additional materials, such as lime or limestone as mentioned above, and the problems of waste control (like disposal of calcium sulfate/sulfite sludge) could prove to be formidable.

One other area which is frequently overlooked in comparing compliance strategies for pollution abatement goals is how well the strategy chosen blends with expected future technolgoy. For example, the electric utility industry is constantly striving for increases in plant efficiency. One of the best or most promising options for improving plant efficiency is referred to as combined-cycle generation. In this concept, fuel is burned in a combustion turbine which drives an electric generator. The hot combustion gas from this turbine is passed to a boiler where steam is generated to drive a steam turbine, which drives another electric generator. It would seem that low-Btu gas is the ideal fuel for combined-cycle generation; undoubtedly, this should create bias in favor of low-Btu gas. There is some possibility that solvent refined coal may prove to be a suitable fuel for combustion turbines; if so, it would then qualify for all the advantages now anticipated for low-Btu gas. In addition, it has the advantage that it can be readily stored and used as needed. This feature is in contrast with the low-Btu gas which must be made practically at the rate at which it is utilized, because storage of large quantities is not feasible.

The influence of the chosen compliance strategy on plant reliability is another most significant consideration. In this instance, both the clean fuel options offer advantages over flue gas cleaning. Clearly, flue gas processing, however reliable it may prove to be, must be performed in series with all other operations in the plant; therefore, the net affect is necessarily to reduce the overall plant reliability. If there is any choice at all between low-Btu gas and solvent refined coal as related to plant reliability, one would assume that the quality of the fuel would weigh less in the consideration than the source of supply. Therefore, since the low-Btu gas plant does not allow the prospect of fuel storage, my own judgment is that it offers somewhat higher risk to reliability than solvent refined coal.

My last item pertains to the sensitivity of the cost of the alternative to the kind of duty cycle which it must perform. To be more specific, the electric utility industry generally classifies facilities in terms of baseload, intermediate load, or peaking load. I suppose that the terminology makes it clear that baseload facilities operate at rated capacity for a large fraction of the time, whereas the peaking facilities are called upon intermittently to meet peak demands. The baseload operation which might be fulfilled in the future with combined-cycle-power plants certainly could be met with either of these three alternatives being discussed, and all three would function better for baseload operation than for either intermediate or peaking duty. On the other hand, as one moves away from baseload operations, it is clear that solvent refining emerges with an increasing advantage because the fuel processing facilities could be operated to give maximum production efficiency and the product could be stored and used as needed. This is not true for low-Btu gas production, and it is presumed that fluctuating demands in flue gas processing would aggravate operational problems which otherwise might not appear.

In summary, the points I have raised in this discussion today are obviously superficial in one sense, but they suggest a line of thought which the power plant operator or planner must follow in order to make rational choices which take into account the constraints I mentioned in the beginning: legal, logistic, technical, and economic. It is perhaps too soon to be very quantitative in analyzing a number of the features of either of the three alternatives which have been used in my remarks, because apparently all three are in their infancy with respect to the requirements which would be imposed on a large commercial venture. As complex and as uncertain as these options may appear, they nevertheless provide a fertile area for analysis and development of system strategies which may be of great value not only to the corporation involved, but to the customers being served.

# ENVIRONMENTAL FACTORS IN COAL LIQUEFACTION PLANT DESIGN

J. B. O'Hara, S. N. Rippee,
B. I. Loran, and W. J. Mindheim*

*Abstract*

*Environmental factors will play an important role in the design and operation of coal liquefaction plants. Such plants are a major national goal. The first large units could be built during this decade. Proposed treatment methods are discussed for solid, liquid, and gaseous effluents based on a preliminary liquefaction plant design developed for the Office of Coal Research by The Ralph M. Parsons Company. An approach to noise control procedures designed to satisfy requirements of the Occupational Health and Safety Act is also described.*

*During the course of future development efforts, further research is recommended to develop additional data and information on environmental factors. Such data will further improve the effectiveness and economy of plant environmental control and of the monitoring systems.*

## INTRODUCTION

Environmental factors were an extremely important influence on the study—recently completed by The Ralph M. Parsons Company for the Office of Coal Research (OCR)—of preliminary design and estimated cost for a demonstration-scale plant to produce clean boiler fuels from coal.

The importance of proper environmental safeguards was considered twofold. First, the design of a demonstration-scale plant is expected to be a forerunner of many large plants producing clean fuels from coal; therefore, the advantages of good environmental protection elements in the design could be magnified many times nationwide. Second, a large body of opinion maintains today that environmental protection and efficient energy production are natural enemies. Since both environmental protection and coal conversion are major national goals, the validity of this widely held opinion would pose a substantial problem to both national programs.

In the Parsons role as technical evaluation contractor to the Office of Coal Research, we found that the environmental protection objectives, as presently known or anticipated by pending legislation, can probably be achieved with appropriate expenditure of money and effort.

Our approach to a discussion and current assessment of environmental factors in coal liquefaction centers on a brief description of the preliminary design for a complex to liquefy 10,000 tons/day of coal. This paper discusses its expected effluent streams and the probable methods of treating them in order to create an environmentally acceptable facility.

The discussion also includes the best judgment estimates of quantities and compositions of certain effluent streams, and our recommendation that coal liquefaction pilot plants emphasize further environmental research and development to insure use of the most effective treatment methods when commercial plants are built.

The design basis of our presentation uses Illinois No. 6 coal to produce two grades of boiler fuels, plus lesser quantities of naphtha and byproduct sulfur. The design was developed as a part of our assignment from the OCR. Reports describing the results have been published by the OCR (ref. 1). Therefore, the process description presented here is brief, and is chiefly intended to show principal sections of the coal conversion complex pertinent to the discussion of environmental factors.

## PROCESS DESCRIPTION

The process configuration is depicted in the block process flow diagram shown in figure 1. This clean boiler-fuel facility consists of a coal preparation section, a coal liquefaction section, and a gasification section. This complex is designed to produce two low-sulfur liquid fuels, sufficient to supply a 600-megawatt power plant. Some naphtha and byproduct sulfur are also produced. The light hydrocarbons formed are burned as plant fuels.

## COAL PREPARATION, DRYING, AND GRINDING

Run-of-the-mine coal is stockpiled and prepared for

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*All are at The Ralph M. Parsons Company, Pasadena, California: O'Hara is Manager of the Energy Department; Rippee is Project Manager; Loran is Senior Environmental Engineer, Systems Division; and Mindheim is Chief Environmental Engineer.

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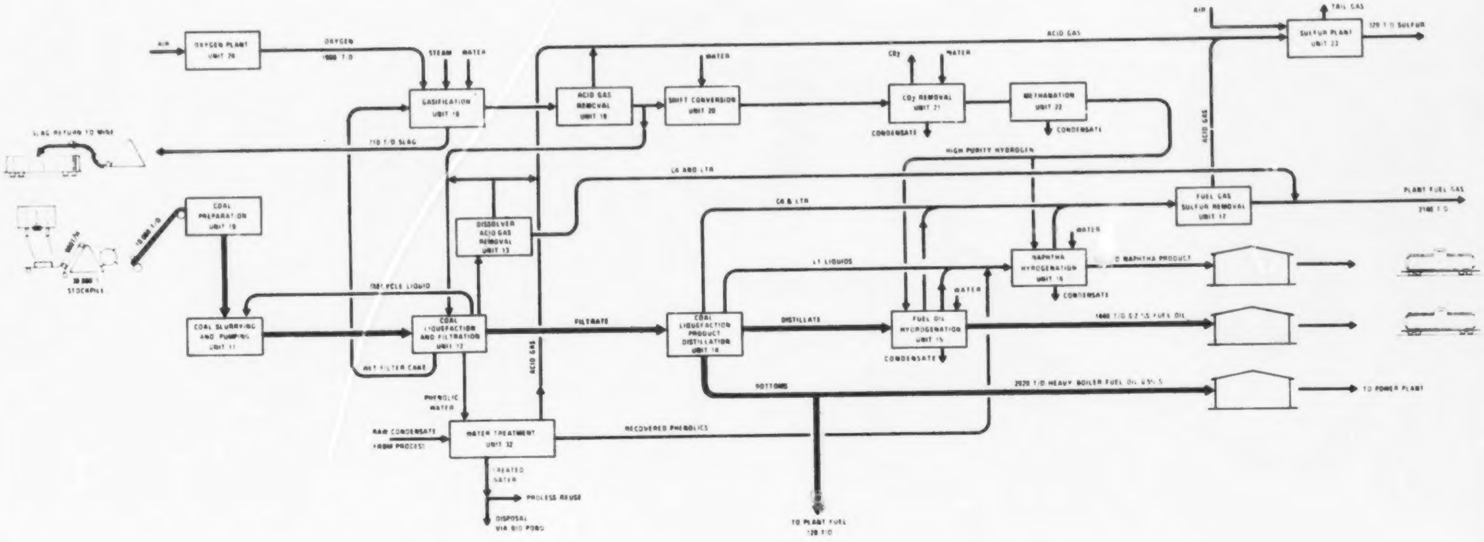

AIR
OXYGEN PLANT UNIT 26
OXYGEN
1800 T/D
STEAM
WATER
GASIFICATION UNIT 19
ACID GAS REMOVAL UNIT 18
WATER
SHIFT CONVERSION UNIT 20
CO2
WATER
CO2 REMOVAL UNIT 21
CONDENSATE
METHANATION UNIT 22
CONDENSATE
ACID GAS
AIR
TAIL GAS
SULFUR PLANT UNIT 23
120 T/D SULFUR
SLAG RETURN TO MINE
710 T/D SLAG
HIGH PURITY HYDROGEN
LG AND LTR
ACID GAS
FUEL GAS SULFUR REMOVAL UNIT 12
PLANT FUEL GAS
2100 T/D
COAL PREPARATION UNIT 10
10 000 T/D
30 000 T
STOCKPILE
DISSOLVER ACID GAS REMOVAL UNIT 13
CG & LTR
WATER
NAPHTHA HYDROGENATION UNIT 16
D NAPHTHA PRODUCT
CONDENSATE
LT LIQUIDS
RECYCLE LIQUID
WATER
COAL SLURRYING AND PUMPING UNIT 11
COAL LIQUEFACTION AND FILTRATION UNIT 12
FILTRATE
COAL LIQUEFACTION PRODUCT DISTILLATION UNIT 14
DISTILLATE
FUEL OIL HYDROGENATION UNIT 15
CONDENSATE
1660 T/D 0.2% S FUEL OIL
WET FILTER CAKE
ACID GAS
BOTTOMS
2020 T/D HEAVY BOILER FUEL OIL 0.5% S
TO POWER PLANT
PHENOLIC WATER
RAW CONDENSATE FROM PROCESS
WATER TREATMENT UNIT 32
RECOVERED PHENOLICS
TREATED WATER
PROCESS REUSE
DISPOSAL VIA BIO POND
TO PLANT FUEL
120 T/D

Figure 1. Demonstration Plant Clean Boiler Fuels From Coal Process Block Flow Diagram

plant feed. Preparation of coal feed consists of a washing plant where a series of jigs, screens, centrifuges, cyclones, and a roll crusher produces washed minus 1¼-inch coal. Refuse from this operation is returned to the mine area for burial. Fine refuse is pumped to settling ponds for further treatment. The crushed coal is then dried in a flow dryer and reduced to minus 1/8 inch in pulverizers for dissolver feed.

## LIQUEFACTION PROCESS

Feed to the liquefaction section consists of minus 1/8-inch coal as a 50-percent-by-weight slurry in a recycle solvent, which is fed to reactors where it is contacted with reducing gases at about 850°F and 1,000 psig. The gas phase of the reactor discharge is largely recycled, while the solid phase is separated from the liquid phase by filtration. The resulting filter cake serves as feed to the gasification section for syngas production.

The liquid phase filtrate produced in the filtration operation is further separated by fractionation into an overhead naphtha stream, a distillate light boiler fuel, and a residual fuel oil. Further hydrogenation of the distillate fuel produces acceptable low-sulfur fuels for boiler firing.

Gases produced in the various units are combined and fed to the acid-gas-removal plant, where carbon dioxide and hydrogen sulfide are removed by scrubbing. The hydrogen sulfide is converted to sulfur in sulfur recovery plants. Carbon dioxide is vented to the atmosphere.

## GASIFIER PLANT

Wet filter cake from the liquefaction process is fed to a slagging, suspension-type gasifier where it reacts with steam and oxygen at 3,000°F and 200 psig pressure. The carbonaceous material is gasified and produces primarily synthesis gas (carbon monoxide and hydrogen). An onsite oxygen plant supplies the required oxygen for this operation.

Most of the cooled syngas is treated for hydrogen sulfide removal and fed directly to the coal liquefaction section of the plant. Syngas not sent directly to the liquefaction section undergoes shift conversion, carbon dioxide removal, and methanation to produce a high purity, hydrogen-gas stream, which is used in hydrogenation of the light distillate and naphtha product streams produced in the liquefaction section.

An overall material balance is shown in figure 2. The 10,000 tons of coal feed are converted into five products. Salable products are 1,440 tons/day of 0.2% sulfur liquid fuel oil, 2,920 tons/day of heavy liquid fuel at 0.5% sulfur, 270 tons/day of naphtha with 1 ppm of sulfur, and 320 tons/day of sulfur. The 2,140 tons/day of plant-produced fuel gas and a small amount of heavy liquid fuel oil are burned for plant operation. The remaining feed streams consist of 1,980 tons/day of oxygen and 21,760 tons/day of water.

Major process waste streams shown consist of 19,430 tons/day of waste gases, 6,390 tons/day of waste water, and a solids waste stream consisting primarily of 710 tons/day of gasifier slag. Each of these categories is discussed in the following sections.

## GASEOUS EFFLUENT

Gaseous process waste streams exhausted to the atmosphere are generated in various sections of the plant. These streams are shown in figure 3, which again depicts the plant process with highlights of gaseous emission streams. Principal gas streams leaving the complex are from the oxygen plant, $CO_2$ removal unit, sulfur plant stack gas, and the combustion gases resulting from fired heaters in the liquefaction and steam generation sections. Off-gases from the gas turbines utilized for power generation are also present.

Figure 4 summarizes the exhaust gas streams and their thermal contents that are expelled from the plant complex. Combustion gases amount to about 90 percent by volume of the total plant gas emission. A total of 990 million cubic feet/day of combustion exhaust gas, containing about 15 billion Btu/day, is exhausted to the atmosphere. Contaminants will consist of sulfur dioxide and nitrogen oxides. When firing with the low-Btu gas produced in the plant, contaminant concentrations will range from about 4 to 15 ppm of sulfur dioxide and 50 to 100 ppm of nitrogen oxides. The small amount of fuel oil required to supplement the low-Btu gas firing for plant power needs contributes about 2.2 million cubic feet/hour of combustion gases containing about 80-100 ppm of sulfur dioxide and 100-150 ppm of nitrogen oxides.

Other waste gases exhausted to the atmosphere consist of the oxygen plant exhaust, sulfur recovery plant tail gas, and the carbon dioxide waste stream from the $CO_2$ removal unit. It would be possible to recover an additional 2,260 tons/day of nitrogen and

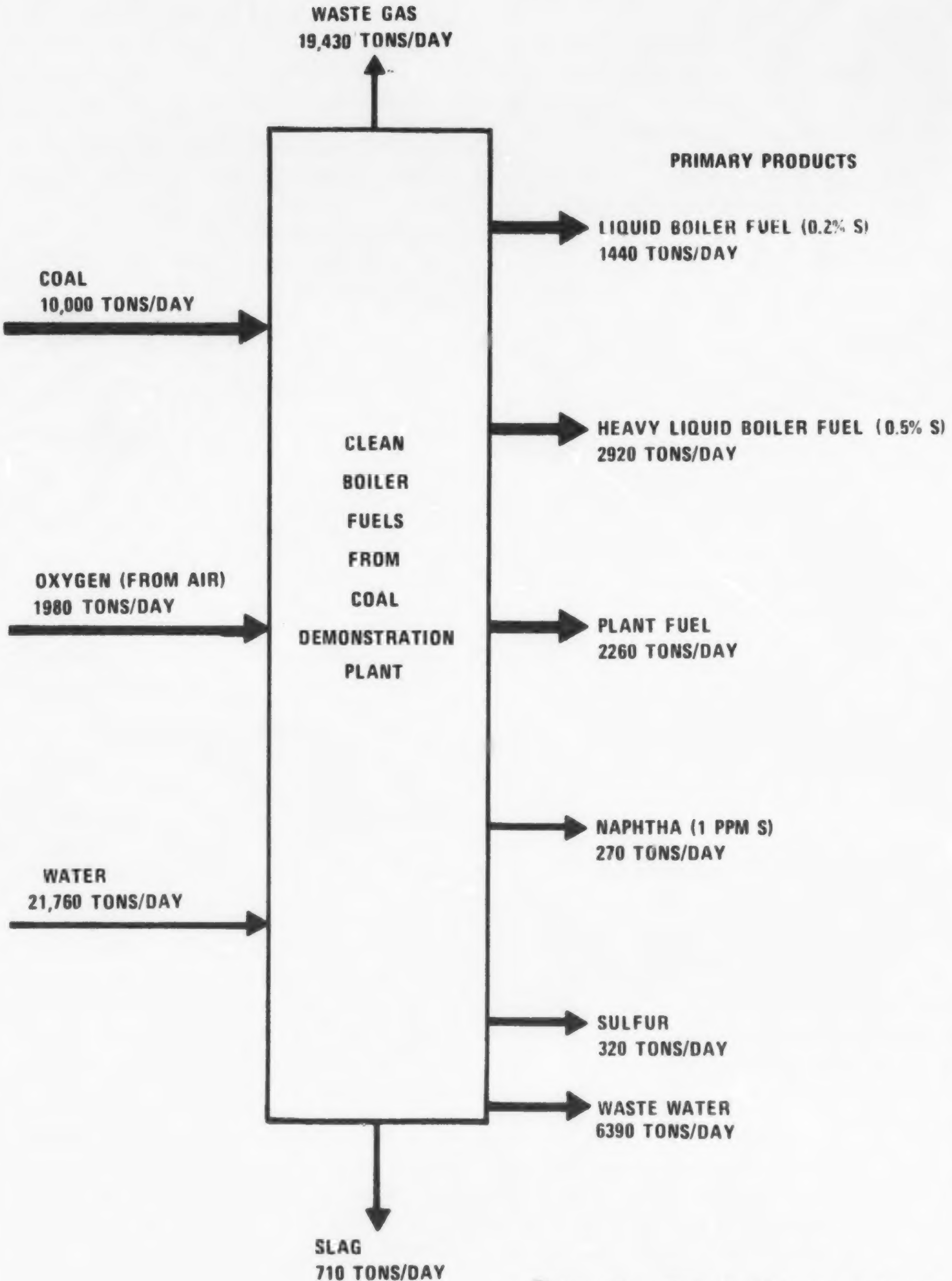

WASTE GAS
19,430 TONS/DAY
PRIMARY PRODUCTS
LIQUID BOILER FUEL (0.2% S)
1440 TONS/DAY
COAL
10,000 TONS/DAY
HEAVY LIQUID BOILER FUEL (0.5% S)
2920 TONS/DAY
CLEAN
BOILER
FUELS
FROM
COAL
DEMONSTRATION
PLANT
OXYGEN (FROM AIR)
1980 TONS/DAY
PLANT FUEL
2260 TONS/DAY
NAPHTHA (1 PPM S)
270 TONS/DAY
WATER
21,760 TONS/DAY
SULFUR
320 TONS/DAY
WASTE WATER
6390 TONS/DAY
SLAG
710 TONS/DAY

Figure 2. Overall Material Balance

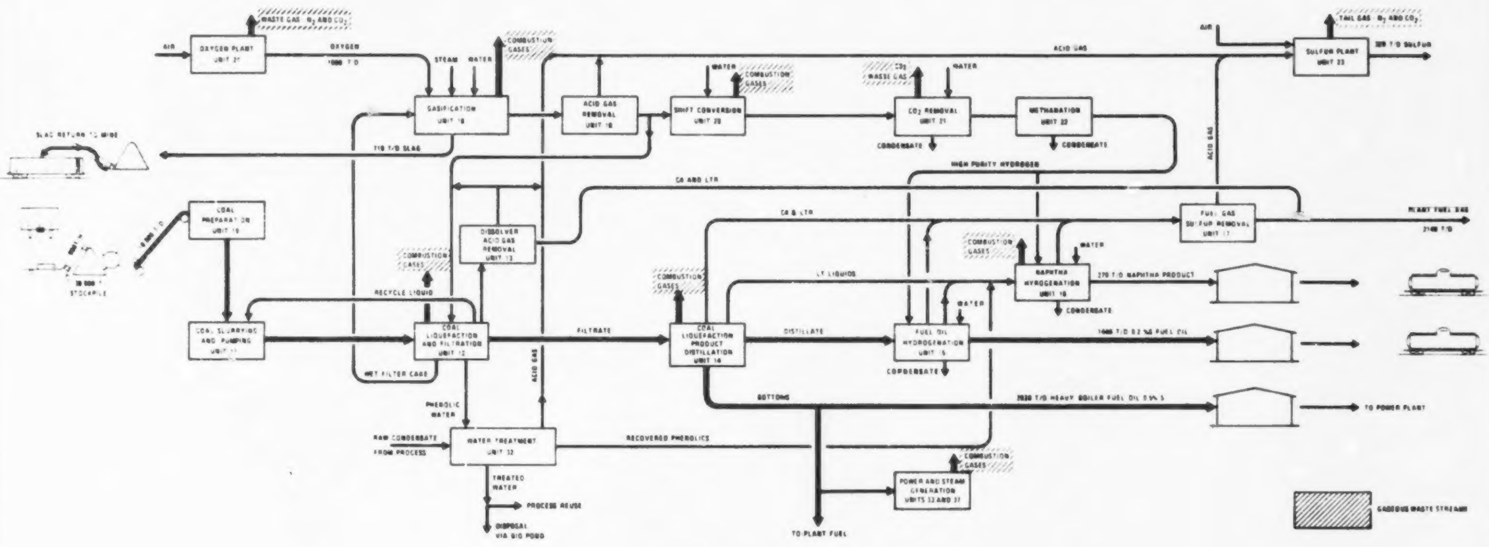
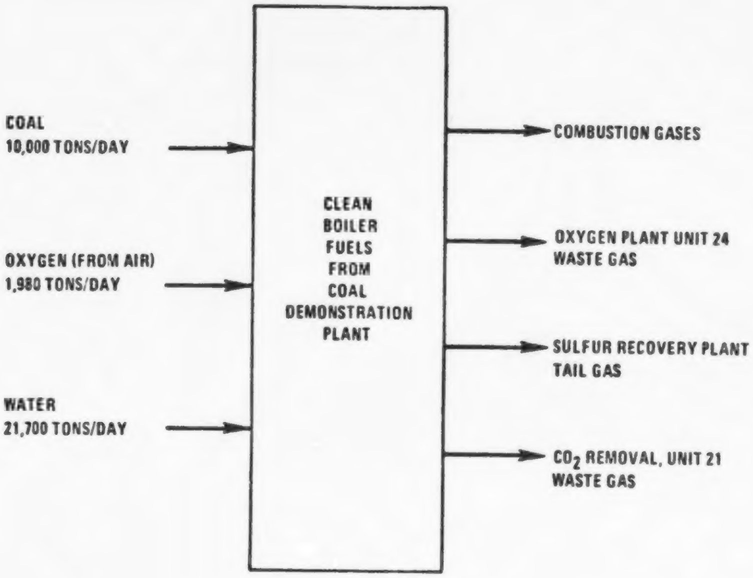

AIR
OXYGEN PLANT
UNIT
WASTE GAS $N_2$ AND $CO_2$
OXYGEN
1000 T/D
STEAM
WATER
COMBUSTION GASES
GASIFICATION
UNIT 18
710 T/D SLAG
SLAG RETURN TO MINE
ACID GAS REMOVAL
UNIT 19
SHIFT CONVERSION
UNIT 20
WATER
COMBUSTION GASES
$CO_2$ WASTE GAS
WATER
$CO_2$ REMOVAL
UNIT 21
CONDENSATE
METHANATION
UNIT 22
CONDENSATE
HIGH PURITY HYDROGEN
ACID GAS
AIR
TAIL GAS $N_2$ AND $CO_2$
SULFUR PLANT
UNIT 23
320 T/D SULFUR
ACID GAS
COAL PREPARATION
UNIT 10
30 000 T
STOCKPILE
COAL SLURRYING AND PUMPING
UNIT 11
RECYCLE LIQUID
COMBUSTION GASES
COAL LIQUEFACTION AND FILTRATION
UNIT 12
WET FILTER CAKE
DISSOLVER ACID GAS REMOVAL
UNIT 13
C6 AND LTR
ACID GAS
FILTRATE
PHENOLIC WATER
RAW CONDENSATE FROM PROCESS
WATER TREATMENT
UNIT 32
TREATED WATER
PROCESS REUSE
DISPOSAL VIA BIO POND
RECOVERED PHENOLICS
COMBUSTION GASES
COAL LIQUEFACTION PRODUCT DISTILLATION
UNIT 14
C6 & LTR
LT LIQUIDS
DISTILLATE
BOTTOMS
TO PLANT FUEL
FUEL OIL HYDROGENATION
UNIT 15
WATER
CONDENSATE
COMBUSTION GASES
NAPHTHA HYDROGENATION
UNIT 16
WATER
CONDENSATE
FUEL GAS SULFUR REMOVAL
UNIT 17
PLANT FUEL GAS
2140 T/D
270 T/D NAPHTHA PRODUCT
1000 T/D 0.2 % S FUEL OIL
2020 T/D HEAVY BOILER FUEL OIL 0.5% S
TO POWER PLANT
POWER AND STEAM GENERATION
UNITS 33 AND 37
COMBUSTION GASES
GASEOUS WASTE STREAM

Figure 3. Demonstration Plant Clean Boiler Fuels From Coal Gaseous Process Waste Streams


COAL
10,000 TONS/DAY
OXYGEN (FROM AIR)
1,980 TONS/DAY
WATER
21,700 TONS/DAY
CLEAN BOILER FUELS FROM COAL DEMONSTRATION PLANT
COMBUSTION GASES
OXYGEN PLANT UNIT 24 WASTE GAS
SULFUR RECOVERY PLANT TAIL GAS
$CO_2$ REMOVAL, UNIT 21 WASTE GAS

| WASTE GAS STREAM | COMPOSITION | OUTPUT, CUBIC FEET/DAY (MILLIONS) | CONTENT, BTU/DAY (MILLIONS) | MAJOR CONTAMINANT |
|---|---|---|---|---|
| COMBUSTION GASES | | | | |
| 1. FUEL GAS | 32% $O_2$, 9.4% $CO_2$, 15.6% $H_2O$, 71.3% $N_2$ | 935 | 14,150 | 4-15 ppm $SO_2$<br>50-100 ppm $NO_x$ |
| 2. FUEL OIL | 3.3% $O_2$, 10.8% $CO_2$, 12.7% $H_2O$, 73.2% $N_2$ | 54 | 850 | 80-100 ppm $SO_2$<br>100-150 ppm $NO_x$<br>35 ppm PARTICULATES |
| OXYGEN PLANT UNIT 24 | $N_2$ AND RARE GASES | 64 | NIL; AMBIENT CONDITIONS | NONE |
| SULFUR REMOVAL PLANT TAIL GAS | 72% $N_2$; REMAINDER $CO_2$ | 32.4 | NIL; AMBIENT CONDITIONS | 10 ppm max $H_2S$ |
| $CO_2$ REMOVAL UNIT 21 | 99% $CO_2$ | 14.3 | NIL; AMBIENT CONDITIONS | NONE |

Figure 4. Gaseous Waste Process Streams

rare gases from the oxygen facilities if area sales justify costs for their recovery. The same is true for the carbon dioxide waste stream from the $CO_2$ removal unit, which amounts to about 14,3 million cubic feet/day of gas containing about 99 percent carbon dioxide.

Waste gas effluent from the sulfur recovery plant would be about 32.4 million cubic feet/day at 93°F. Hydrogen sulfide content of this stream during normal operation is expected to be less than 5 ppm. The main constituent of the gas stream is 72 percent nitrogen. The remainder is primarily carbon dioxide.

In summary, approximately 1 billion cubic feet/day of various gas streams from the complex are exhausted to the atmosphere. All gaseous waste effluent streams meet applicable standards. Fuel combustion gases exhausted will meet ambient air quality standards for nitric oxide ($NO_x$) and sulfur dioxide ($SO_2$); this is also true for particulate (fly ash) entrained in the gases since primarily gaseous fuels are used. When fuel oil is used to supplement gaseous fuel, particulates are estimated at about 39 ppm, which meets ambient air quality standards. The rate of $NO_x$ production by various plant fuel sections is a function of maximum flame temperature and retention time. Furnace designs shall be such that exit combustion gases shall meet the nitrogen dioxide ($NO_2$) standards. The gas waste effluent stream from the sulfur recovery unit is designed to operate normally at about 5 ppm of hydrogen sulfide, well within standards.

## LIQUID EFFLUENT

Figure 5 is a flow diagram showing the major aqueous wastewater streams leaving the plant complex. Approximately 532,000 pounds/hour of wastewater are discharged from the complex, or about 1,060 gallons/minute. Cooling tower blowdown is slightly more than one-half of the total, or about 600 gallons/minute. Sanitary waste water, boiler blowdown, treated oily water, and stripped plant sour water make up the remainder of the plant complex wastewater stream.

Process means have been provided for stripping nonphenolic process water, and this stripped water is returned to process for reuse. Also, stripped waste phenolic water effluent, which contains the greatest number of pollutants, will average about 40 gallons/minute. This stream joins the sanitary waste, treated oily water, cooling tower, boiler blowdown streams, and backwash water from demineralizers and sand filters. This combined stream undergoes final treatment in the aerated lagoon and biopond before leaving the complex.

Cooling tower and boiler blowdown streams are expected to contain not greater than 15 ppm of phosphate, 10 ppm of chromate, and 5 ppm of zinc.

Table 1 summarizes the estimated wastewater treatment data and contaminants in the effluent stream leaving the complex.

The total oil in the process waste stream is expected to consist of 80 percent by weight of naphtha and 20 percent acid oil, which amounts to about 5 ppm TOC feed. The COD feed is estimated at a level of about 150 ppm with a BOD level of about 40 ppm.

The aerated lagoon operation is expected to provide reduction of 96 percent of the inlet sulfide concentration, 94 percent of ammonia, 88 percent of acid oil, 75 percent of BOD, 75 percent of suspended solids, 95 percent of phenols, 69 percent of COD, and 99 percent of phosphates. The aerated lagoon is expected to handle most of the impurities in the waste streams of the coal conversion complex based on a flow of water from the phenolic sour water stripper of 40 gallons/minute into the aerated lagoon. However, it is possible that phenol and heavy metals may still exceed local or State standards. In this case, they will be further reduced by adding an activated sludge plant prior to final biopond streatment or by extending the retention time and providing more aeration to the biopond. The final design decision will be based upon data obtained from the pilot plants.

Dissolved impurities from the cooling tower and boiler blowdown streams are stable and will not be destroyed by either impounding or aeration. However, it is possible to eliminate chromates by utilizing organic and biodegradable cooling water inhibitors. These inhibitors are normally not as economical or effective as chromates. Provision has been made for pretreatment of these streams in the neutralization pit, should it be necessary to precipitate impurities prior to pumping to the aerated lagoon.

## SOLID WASTES

The block flow diagram of figure 6 shows the types of waste solids generated by the complex. Major solid wastes are produced during pretreatment of the run-of-mine coal and gasification of the liquefaction filter cake to produce syngas. Additional solids of lesser quantity also requiring disposal are various

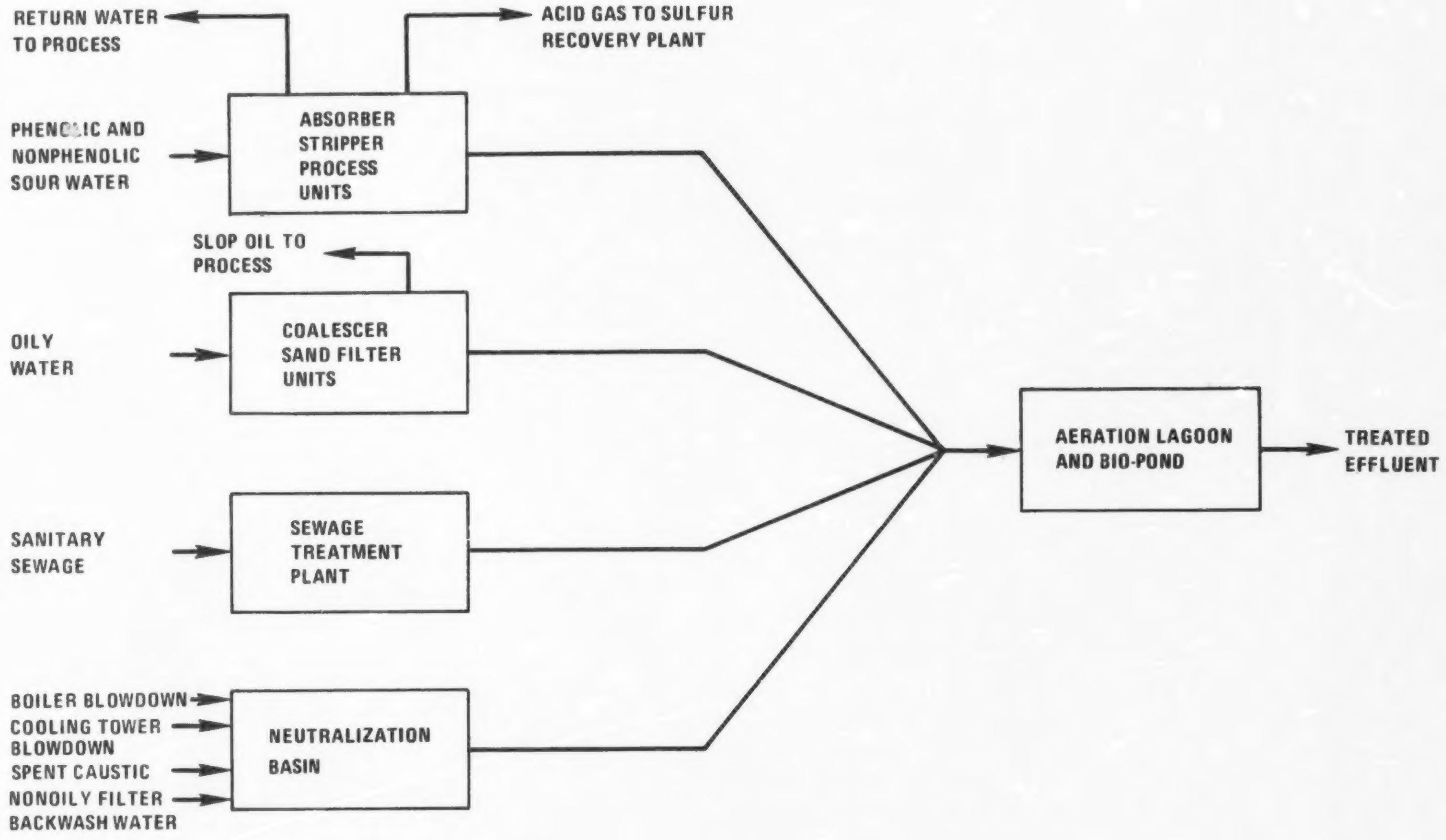

RETURN WATER TO PROCESS
ACID GAS TO SULFUR RECOVERY PLANT
PHENOLIC AND NONPHENOLIC SOUR WATER
ABSORBER STRIPPER PROCESS UNITS
SLOP OIL TO PROCESS
OILY WATER
COALESCER SAND FILTER UNITS
SANITARY SEWAGE
SEWAGE TREATMENT PLANT
BOILER BLOWDOWN
COOLING TOWER BLOWDOWN
SPENT CAUSTIC
NONOILY FILTER BACKWASH WATER
NEUTRALIZATION BASIN
AERATION LAGOON AND BIO-POND
TREATED EFFLUENT

Figure 5. Demonstration Plant—Clean Boiler Fuels From Coal Major Aqueous Waste Water Streams

Table 1. Estimated waste bio-pond effluent concentrations for demonstration plant

| CONSTITUENTS | BIOLOGICAL CHARGE | | BIOLOGICAL EFFLUENT | | STATE REQUIREMENT |
|---|---|---|---|---|---|
| | LB/DAY | PPM | LB/DAY | PPM | PPM |
| SULFIDE | 1.48 | 0.12 | 0.06 | 0 | – |
| AMMONIA | 24 | 1.88 | 1.45 | 0.11 | 2.5 |
| OIL | 72 | 5.63 | 8.64 | 0.68 | 1.0 |
| TOC | 60 | 4.69 | – | – | – |
| $BOD_5$ | 538 | 42 | 134.5 | 10.5 | 20 |
| SS | 660 | 51.6 | 165 | 12.9 | 25 |
| PHENOL | 96 | 7.5 | 4.8 | 0.38 | 0.3 |
| COD | 1,920 | 150 | 576 | 45 | – |
| PHOSPHATE | 145 | 11.3 | 1.45 | 0.11 | 1.0 as P |
| PH | 6-9 | 6-9 | 6-9 | 6-9 | 5-10 |
| CHROMATE | 91 | 7.1 | 91 | 7.1 | 0.05 as $CrO_4$ |
| ZINC | 45 | 3.5 | 45 | 3.5 | 1.0 |
| COLIFORM ORGANISM | | 15/100 ml | | 15/100 ml | 400/100 ml |

TOTAL ESTIMATED FLOW TO BIO-POND
1060 GAL/MIN ≈ 12.8 MILLION LB/DAY

spent catalysts generated in the hydrogenation, shift reaction, methanation, and tail gas sections. After being rendered inert, these spent catalysts, which have a lifetime of 2 to 3 years, will be disposed of by backhaul and will fill in the mined-out coal areas.

In the coal preparation plant, reject material from the primary coal breaker amounts to about 450 tons/hour. This material is combined with the double-deck screen reject, about 88 tons/hour, and conveyed to a truck-loading bin. Trucks transport the

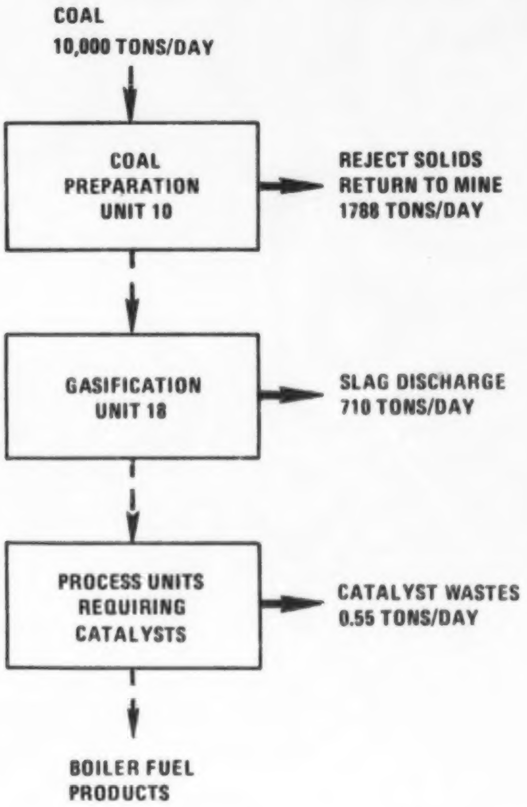

COAL
10,000 TONS/DAY
COAL PREPARATION UNIT 10
REJECT SOLIDS RETURN TO MINE 1788 TONS/DAY
GASIFICATION UNIT 18
SLAG DISCHARGE 710 TONS/DAY
PROCESS UNITS REQUIRING CATALYSTS
CATALYST WASTES 0.55 TONS/DAY
BOILER FUEL PRODUCTS

Figure 6. Demonstration plant—clean boiler fuels from coal major solid waste streams

waste material to the coal mine for burial in mined-out areas. This solid consists mainly of low-grade coal and shale and is about minus 3 inches in size. The thickener underflow, which is a fine reject material of minus 1/16 inch in size, is pumped to a tailings-pond for solids recovery and for recycle of the decanted water to the coal preparation plant These solids also consist primarily of low-grade coal and shale.

Gasifier slag produced is approximately 710 tons/day. This material will be dewatered and conveyed to a truck-loading bin for transport to the mined-out areas of the coal mine for burial. The material will probably be utilized later as an inert additive in the manufacture of cinder blocks.

## NOISE POLLUTION

Figure 7 shows possible plant and equipment areas suspected of noise pollution. The Occupational Safety and Health Act of 1970 regulates the amount of "weighted" noise to which a worker can be exposed, in order to protect him from ear damage. Local code usually regulates the amount of noise, in decibels, that an industrial plant can generate above the normal ambient background level of the community, measured at the property line. Noise control is an integral part of the layout and design of coal conversion plants. During equipment design and engineering layout, special attention will be given to fans and compressors, gasifiers, fired heaters, and gas turbine areas to minimize noise-source levels and any excessive noise radiation effect on plant personnel.

Equipment vendors will be requested to show evidence that installed equipment will meet noise level requirements. However, noise from equipment components may not represent the total sound level, including all equipment items, motor drives, piping or ductwork, and other associated equipment. Added to these factors are reverberations from adjacent equipment, buildings, and sound interferences from different sound sources. Consequently, total engineering plant and equipment layout design will play an important part in lessening plant noise level.

## DEVELOPMENT PROGRAM RECOMMENDATIONS

Past experience and judgment superimposed on the preceding brief summary and analysis, led us to recommend that further coal liquefaction and gasification research is required to determine the presence or absence of the constituents shown in table 2. If present, they are expected to occur in minor concentrations, but will require proper treatment facilities. Availability of this added information will further improve reliability of the plant's environmental control and monitoring systems.

## REFERENCE

1. *Demonstration Plant - Clean Boiler Fuels From Coal - Preliminary Design/Capital Cost Estimate*, R & D Report No. 82, Interim Report No. 1, Vols. I and II, prepared by The Ralph M. Parsons Company for the United States Department of the Interior, Office of Coal Research.

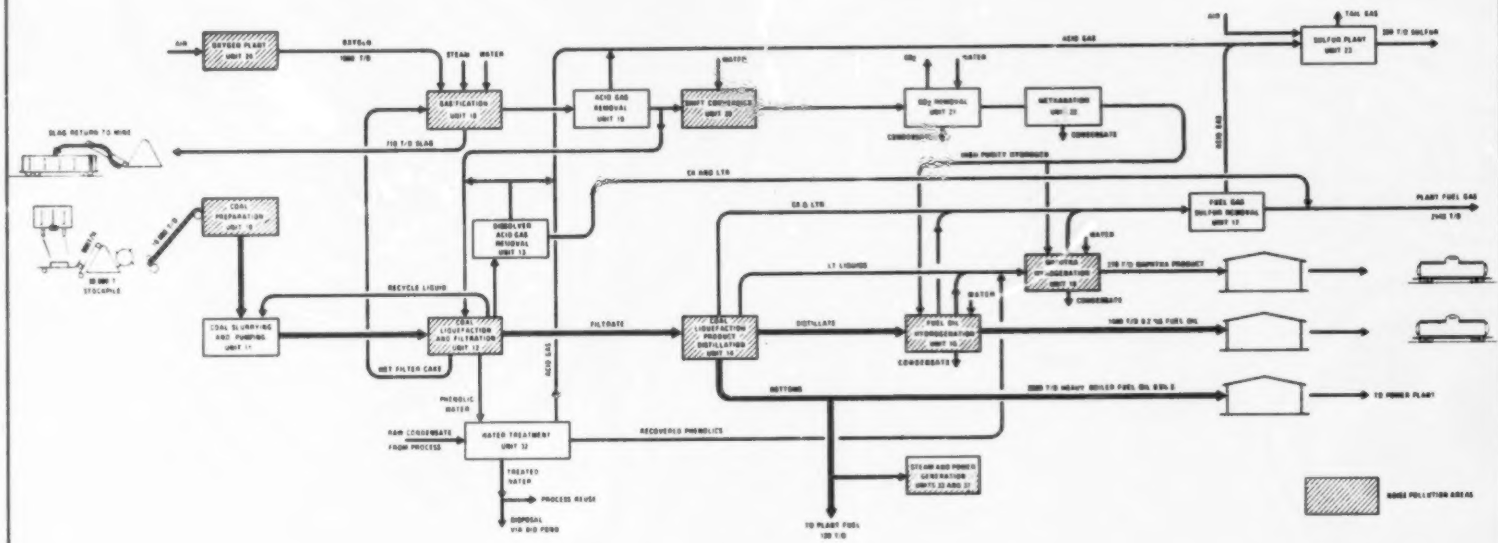

AIR
OXYGEN PLANT UNIT 20
OXYGEN 1000 T/D
STEAM
WATER
GASIFICATION UNIT 18
ACID GAS REMOVAL UNIT 19
SHIFT CONVERSION UNIT 20
WATER
CO2
WATER
CO2 REMOVAL UNIT 21
CONDENSATE
METHANATION UNIT 22
CONDENSATE
ACID GAS
AIR
TAIL GAS
SULFUR PLANT UNIT 23
200 T/D SULFUR
SLAG RETURN TO MINE
110 T/D SLAG
CH AND LTR
HIGH PURITY HYDROGEN
ACID GAS
COAL PREPARATION UNIT 10
STOCKPILE
DISSOLVER ACID GAS REMOVAL UNIT 13
CH & LTR
FUEL GAS SULFUR REMOVAL UNIT 17
PLANT FUEL GAS
2140 T/D
WATER
LT LIQUIDS
NAPHTHA HYDROGENATION UNIT 18
270 T/D NAPHTHA PRODUCT
CONDENSATE
RECYCLE LIQUID
COAL SLURRYING AND PUMPING UNIT 11
COAL LIQUEFACTION AND FILTRATION UNIT 12
FILTRATE
COAL LIQUEFACTION PRODUCT DISTILLATION UNIT 14
DISTILLATE
WATER
FUEL OIL HYDROGENATION UNIT 15
1000 T/D 0.2 % S FUEL OIL
CONDENSATE
WET FILTER CAKE
ACID GAS
BOTTOMS
2000 T/D HEAVY BOILER FUEL OIL 0.5% S
TO POWER PLANT
PHENOLIC WATER
RAW CONDENSATE FROM PROCESS
WATER TREATMENT UNIT 32
RECOVERED PHENOLICS
TREATED WATER
PROCESS REUSE
DISPOSAL VIA BIO POND
STEAM AND POWER GENERATION UNITS 33 AND 31
TO PLANT FUEL 120 T/D
NOISE POLLUTION AREAS

Figure 7. Demonstration Plant—Clean Boiler Fuels From Coal Unit Areas Possible Noise Pollution

Table 2. Recommended additional contaminant research program for coal liquefaction plants

| COMPOUND | EXPECTED TO OCCUR IN | REMARKS |
|---|---|---|
| AMMONIA ($NH_3$) | BOTH GASIFICATION AND LIQUEFACTION | ASSUME PRESENT; QUANTITY NEEDS VERIFICATION |
| HYDROGEN CYANIDE | BOTH GASIFICATION AND LIQUEFACTION | ASSUME PRESENT; QUANTITY NEEDS VERIFICATION |
| THIOCYANATES | BOTH GASIFICATION AND LIQUEFACTION | PRESENCE SUSPECTED; NEED DATA |
| PHENOLS | LIQUEFACTION | PRESENT; QUANTITY NEEDS VERIFICATION |
| ORGANIC ACIDS ALDEHYDES AND KETONES | LIQUEFACTION | PRESENT; QUANTITY NEEDS VERIFICATION |
| METAL SULFIDES | LIQUEFACTION | PRESENCE SUSPECTED; NEED DATA |
| MERCAPTANS | LIQUEFACTION | PRESENCE SUSPECTED; NEED DATA |
| CARBON DISULFIDE<br>CARBONYL SULFIDE | BOTH GASIFICATION AND LIQUEFACTION | PRESENT; REMOVED IN GAS PURIFICATION STEPS; QUANTITY NEEDS VERIFICATION |
| COAL-TRACE ELEMENTS (Be, F, As, Hg, AND Pb) | BOTH GASIFICATION AND LIQUEFACTION | GASIFICATION DATA AVAILABLE INDICATES PRESENCE; LIMITED DATA AVAILABLE ON LIQUEFACTION PROCESS |

# COLONY
# OIL SHALE DEVELOPMENT
# PARACHUTE CREEK, COLORADO

Mark T. Atwood*

*Abstract*

*Production of oil from oil shale involves mining, crushing, retorting (pyrolysis), and product oil upgrading. The disposal of the processed shale and vegetation of the exposed surface is a major environmental consideration. Several years of intensive investigation have provided satisfactory solutions to all problem areas.*

## BACKGROUND

Work on the development of the TOSCO II oil-shale-retorting-process was begun in 1956 by The Oil Shale Corporation. A pilot plant was built in 1957 near Littleton, Colorado. All of the early experimental work was contracted to the Denver Research Institute.

In 1964, Colony Development Company (Agent) was formed by Standard Oil Company of Ohio, The Cleveland-Cliffs Iron Company, and The Oil Shale Corporation (TOSCO). In the following year, this group began operation of a 1,000-ton-per-day semiworks retort and mining operation at a site 17 miles north of Grand Valley, Colorado. The Oil Shale Corporation continued operations on its own in 1966 and 1967; in 1969, Atlantic Richfield Company joined Colony as operator.

Semiworks testing was intensified in 1971 and continued into April 1972, at which time operations were suspended and the emphasis was shifted to commercial design and environmental studies. In 1973, TOSCO and Atlantic Richfield selected C. F. Braun & Co. as managing contractor for a 45,000-barrel-per-day commercial oil shale complex to be constructed on Colony's Dow property at Parachute Creek.

On January 5, 1974, Ashland Oil, Inc., joined the commercialization program. On February 11, 1974, Shell Oil Company signed a letter of intent to participate in the preconstruction program. Field construction is expected to begin in the fall of 1974 or early 1975 if necessary government permits are received and adequate financial arrangements are completed in time.

## THE TOSCO II PROCESS

A schematic of the TOSCO II retort is shown in figure 1. Raw shale is fed through a surge hopper and preheated by dilute phase fluid bed techniques. The preheated feed is then transported to a pyrolysis drum where it is contacted with heated ceramic pellets. The solid, processed shale leaves the pyrolysis drum and passes through the trommel screen; then it is cooled and goes to storage. The cooled ceramic pellets pass over the trommel screen and return to the pellet heater by the ball elevator. Pyrolysis vapors are condensed in the fractionator. Uncondensed hydrocarbon gases can be utilized as an inplant fuel, as indicated in figure 1, or can be processed to produce hydrogen.

Since the retorting step itself is carried out with an externally heated transfer medium, the pyrolysis gases are obtained undiluted with nitrogen and combustion derived carbon dioxide. A typical TOSCO II process gas analysis is given in table 1. Typical shale oil from the TOSCO II process is described in table 2.

## PARACHUTE CREEK FUTURE COMMERCIAL OPERATIONS

The location of the Dow property and the areas of major development within it are shown in figure 2. A mine bench is located at the base of Middle Fork of Parachute Creek and the latest planned plant site is on the top of the Roan Plateau. Processed shale disposal will be as indicated in Davis Gulch.

A schematic diagram of the oil shale processing operation is shown in figure 3. This diagram illustrates the consecutive steps of mining, crushing, stockpiling, final crushing, pyrolysis, and spent shale disposal. It also shows the facilities for upgrading the raw shale oil to a final product containing very low levels of nitrogen and sulfur. This final product would serve as an environmentally acceptable fuel oil.

Mining will be conducted by conventional room and pillar techniques, as illustrated by figure 4. Two 30-foot benches will be mined by the sequence of drilling, emplacement of explosives, blasting, and mucking. Front end loaders will place the broken shale rock into trucks for movement to the primary crusher. The top bench will be extracted before

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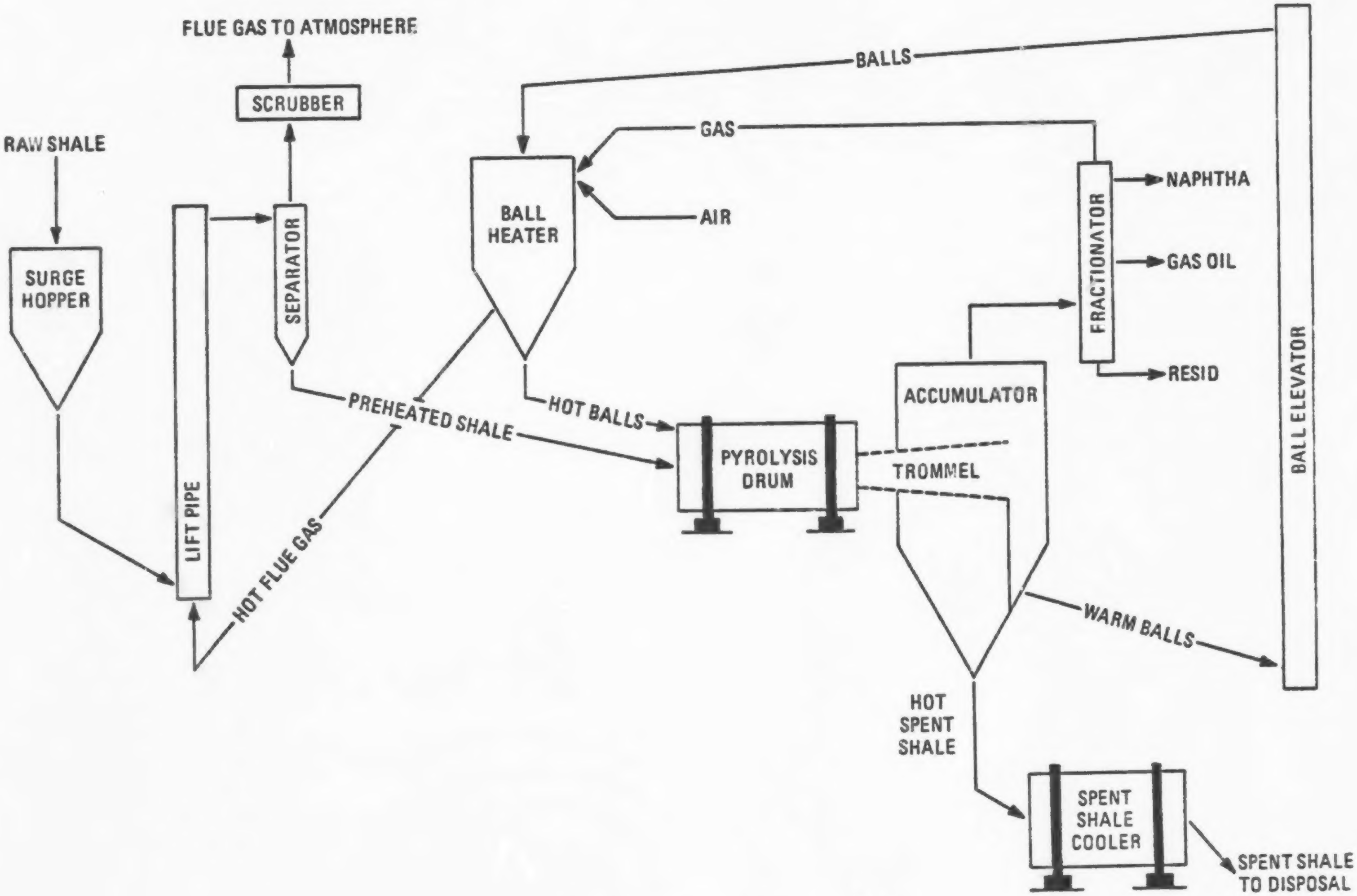

FLUE GAS TO ATMOSPHERE
SCRUBBER
RAW SHALE
SURGE HOPPER
LIFT PIPE
SEPARATOR
HOT FLUE GAS
PREHEATED SHALE
BALLS
GAS
AIR
BALL HEATER
HOT BALLS
PYROLYSIS DRUM
TROMMEL
ACCUMULATOR
FRACTIONATOR
NAPHTHA
GAS OIL
RESID
BALL ELEVATOR
WARM BALLS
HOT SPENT SHALE
SPENT SHALE COOLER
SPENT SHALE TO DISPOSAL

Figure 1. Topco II Process

Table 1. TOSCO II process typical gas analyses

(33 gallons per ton raw shale)

| Component | Weight percent | Mole percent |
|---|---|---|
| $H_2$ | 1.53 | 22.44 |
| CO | 3.37 | 3.56 |
| $CH_4$ | 8.25 | 15.23 |
| $C_2H_6$ | 10.53 | 10.31 |
| $C_2H_4$ | 5.07 | 5.36 |
| $C_3H_8$ | 6.00 | 4.03 |
| $C_3H_6$ | 5.25 | 3.71 |
| $i\text{-}C_4H_{10}$ | 0.49 | 0.26 |
| $n\text{-}C_4H_{10}$ | 2.54 | 1.30 |
| Butenes | 5.11 | 2.70 |
| $C_5$'s | 6.18 | 2.55 |
| $C_6$'s | 4.35 | 1.54 |
| $C_7$'s | 2.81 | 0.83 |
| $C_8$'s | 1.35 | 0.36 |
| $C_8^+$ | 0.36 | 0.09 |
| $CO_2$ | 31.84 | 21.43 |
| $H_2S$ | 4.97 | 4.30 |
|  | 100.00 | 100.00 |

Table 2. Typical shale oil from the TOSCO II process

| | |
|---|---|
| Gravity, °API | 22.0 |
| Pour point, (°F) | 30.0 |
| Sulfur, (wt %) | 0.8 |
| Nitrogen, (wt %) | 1.8 |
| Carbon, (wt %) | 84.7 |
| Hydrogen, (wt %) | 11.3 |
| Carbon to hydrogen, weight ratio | 7.5 |
| **Distillation** | **Volume percent** |
| IBP to 400°F | 18.0 |
| 400 to 600°F | 24.0 |
| 600 to 900°F | 34.0 |
| 900°F and heavier | 24.0 |

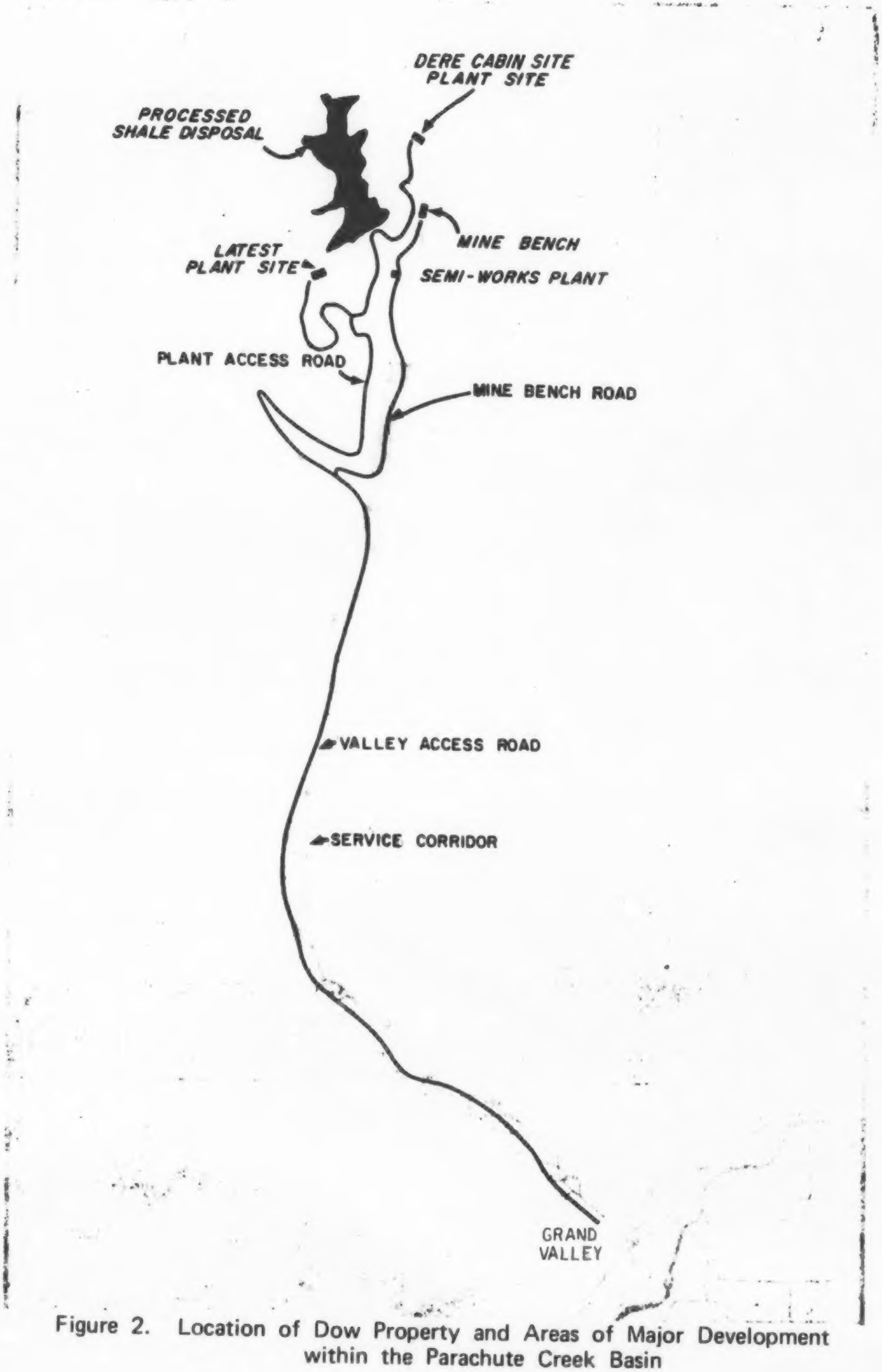

DERE CABIN SITE
PLANT SITE
PROCESSED
SHALE DISPOSAL
MINE BENCH
LATEST
PLANT SITE
SEMI-WORKS PLANT
PLANT ACCESS ROAD
MINE BENCH ROAD
VALLEY ACCESS ROAD
SERVICE CORRIDOR
GRAND
VALLEY

Figure 2. Location of Dow Property and Areas of Major Development within the Parachute Creek Basin

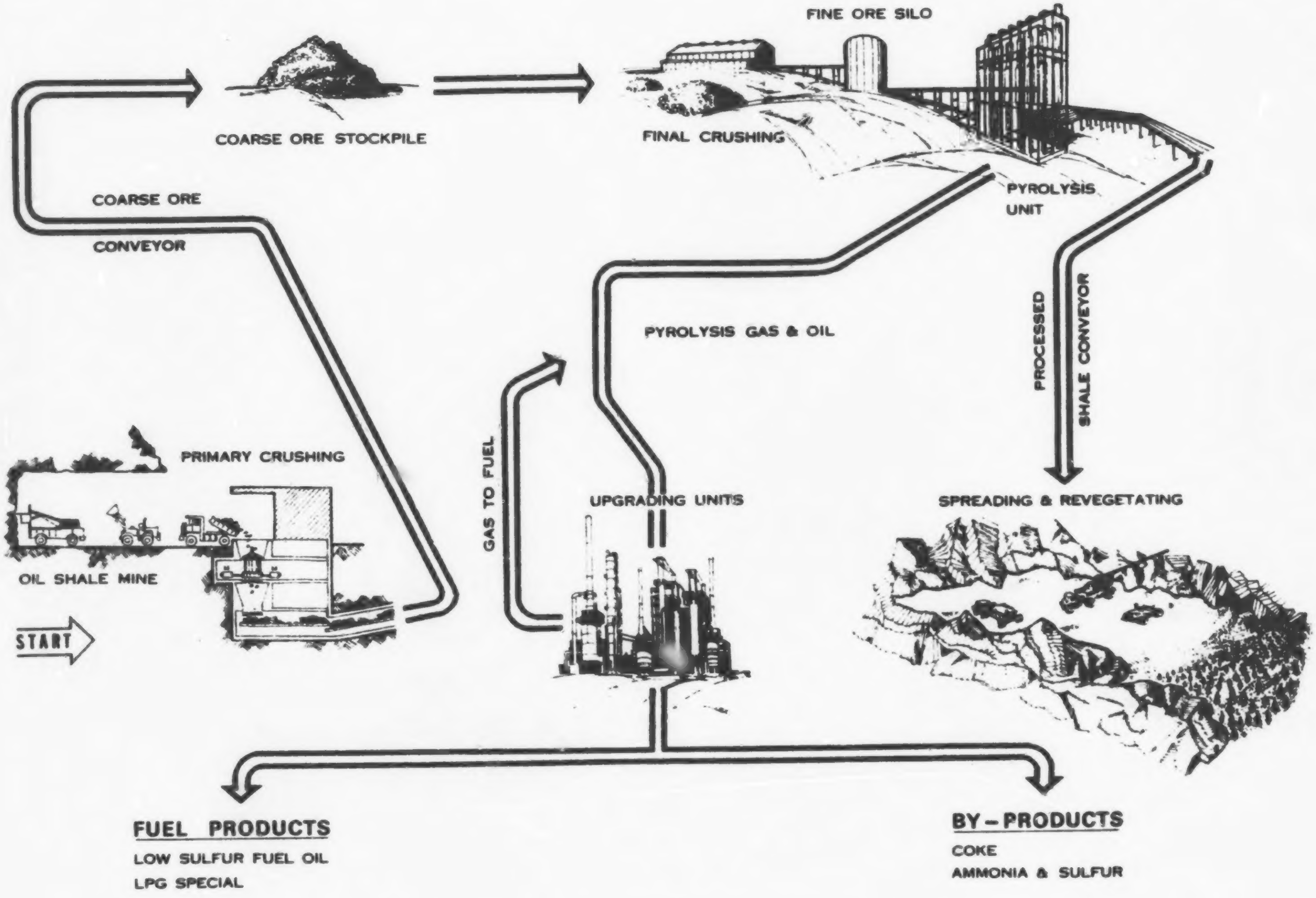

FINE ORE SILO
COARSE ORE STOCKPILE
FINAL CRUSHING
COARSE ORE
CONVEYOR
PYROLYSIS UNIT
PYROLYSIS GAS & OIL
PROCESSED
SHALE CONVEYOR
PRIMARY CRUSHING
GAS TO FUEL
UPGRADING UNITS
SPREADING & REVEGETATING
OIL SHALE MINE
START
FUEL PRODUCTS
LOW SULFUR FUEL OIL
LPG SPECIAL
BY-PRODUCTS
COKE
AMMONIA & SULFUR

Figure 3. Schematic Diagram -- Oil Shale Processing

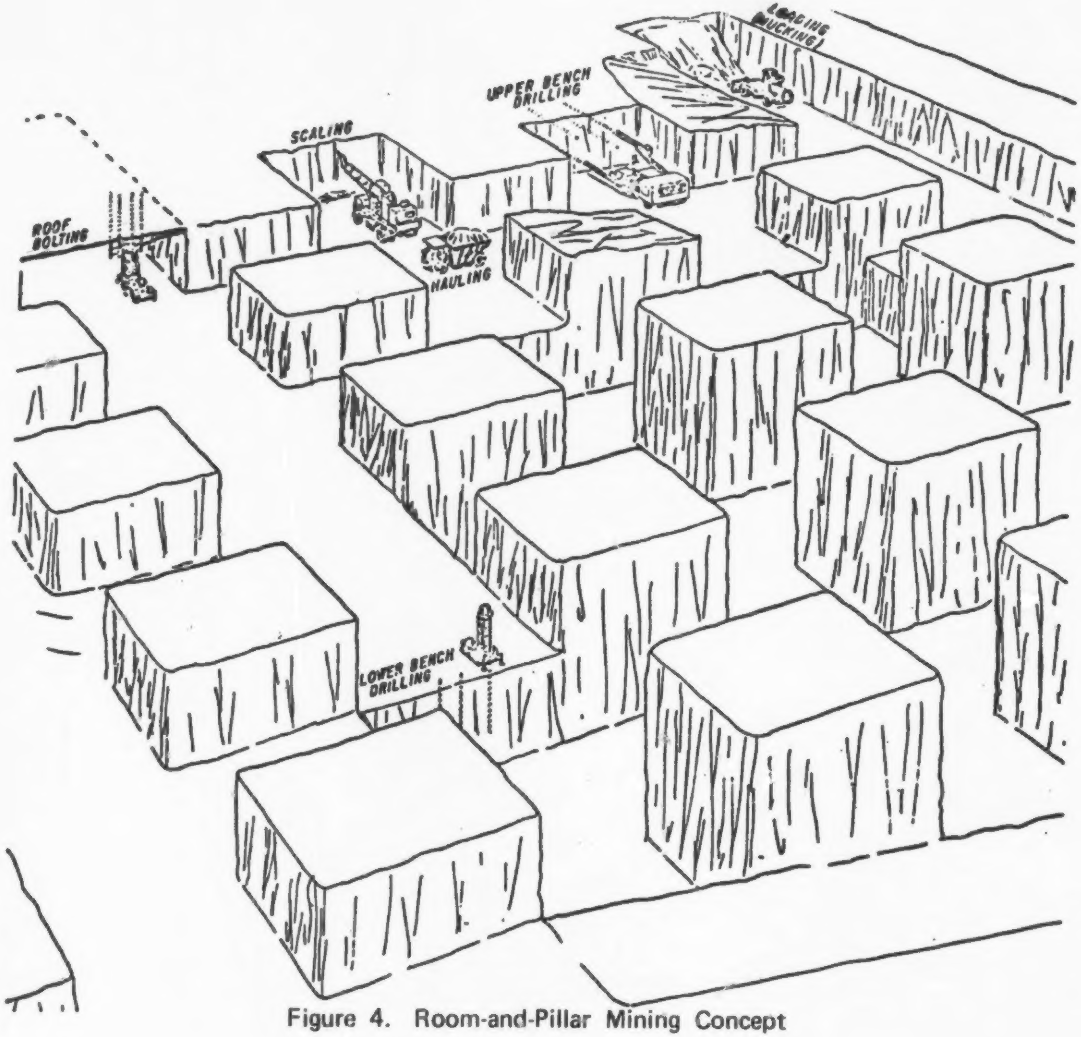

LOADING (MUCKING)
UPPER BENCH DRILLING
SCALING
ROOF BOLTING
HAULING
LOWER BENCH DRILLING

Figure 4. Room-and-Pillar Mining Concept

mining the lower bench. Approximately 61,000 tons will be mined per day. About 60 percent of the inplace shale will be removed with the remainder to be left in place as pillars to support the overburden. Eventually, 4,100 acres will be mined in this fashion on the western portion of the Dow property.

Figure 5 shows the shale-oil-complex site indicating positions of the mine, processing, and processed shale disposal areas.

A closeup of the retorting and upgrading units on the plateau is shown in figure 6. This sketch illustrates the movement of coarse shale to the secondary crushing operation and the six pyrolysis units, each having a capacity of around 11,000 tons per day. In the foreground is the upgrading or hydrotreating facilities which produce the final product oil. Byproducts produced are sulfur, coke, and ammonia.

Construction of Colony's commercial oil shale complex at Parachute Creek will require about 40 months and a peak construction force of about 1,200 employees.

## PROCESSED SHALE DISPOSAL

The spent shale remaining after pyrolysis in a TOSCO II retort has been termed "processed shale" (ref. 1). It is a dark, granular solid which contains residual carbon but no residual kerogen.

About 400 million tons of this processed shale will be disposed of in Davis Gulch during 20 years of commercial operation. This particular small valley has a very limited watershed and low stream flow, and the flooding hazard will be minimal. The moistened processed shale will be deposited initially in side draws of Davis Gulch to eliminate contact with natural aquifers. Eventually, 800 acres will be covered with the processed shale.

After compaction, the processed shale is nearly impermeable and will not permit leaching or percolation of water into surrounding aquifers. Engineering studies indicate that a 1:3 slope (vertical:horizontal) on the face is adequate from a safety point of view, and Colony plans to use a slope of less than 1:4 as an additional safety factor. Due to the possibility of surface erosion from the action of rain and melting snow, the surface will be benched at regular intervals. This will reduce surface water flow velocities and allow any sediment being carried to settle out. This configuration is illustrated in figure 7.

As indicated in figure 7, a detention structure will be built upstream of the processed shale pile to prevent ground water from reaching the pile, and a catchment dam will be placed downstream of the pile to collect any surface runoff. The surface runoff will be returned to the processed shale moisturizer and thus eventually returned to the processed shale pile.

As permanent surfaces are created in the processed shale placement operation, revegetation will begin at the first available planning time in order to control wind and water erosion. Prior to commencement of revegetation, water spraying will be utilized to control any dusting.

## PROCESSED SHALE REVEGETATION

Work has been conducted at Colony Development since 1965 with two major goals: (1) to establish a long-lasting cover as soon as possible, and (2) to obtain an ecosystem which closely resembles that presently existing on the plateau (ref. 2).

The characteristics of freshly processed shale, as compacted in the disposal area, which will govern revegetation are as follows:

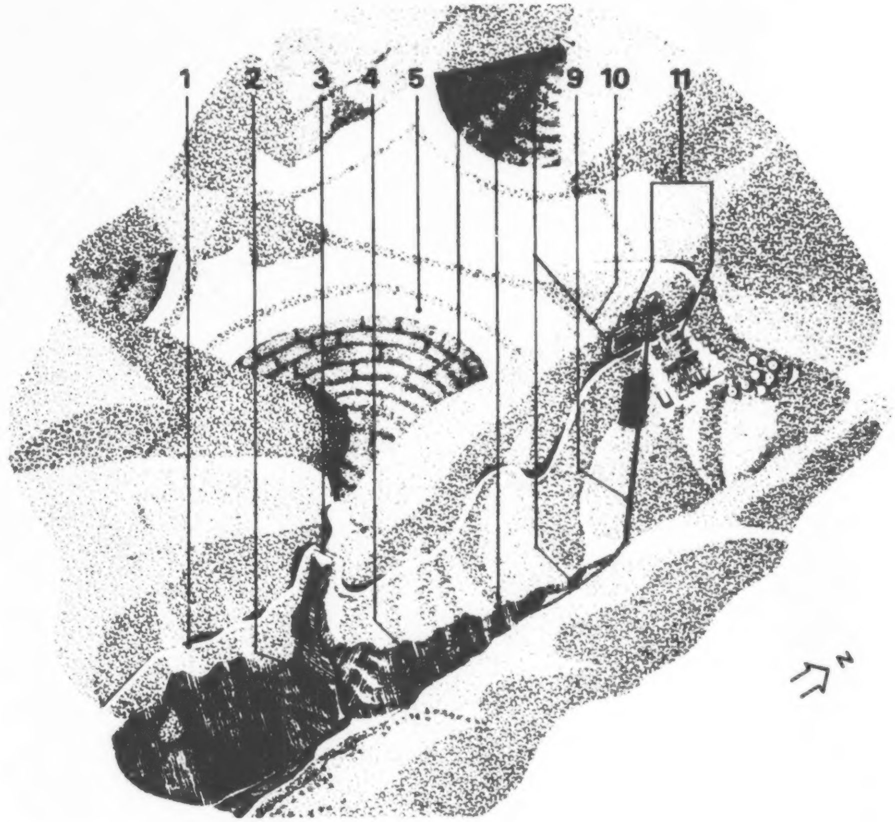
1. Low amount of available nutrients;
2. High soluble salt content;
3. Structural characteristics which, after compaction, render it essentially impermeable to precipitation.

The initial low fertility will be improved by the application of fertilizers. Current plans are to add 600 pounds of phosphorus and 70 pounds of nitrogen per acre in initial applications. Later, it may be desirable to add organic matter, such as sewage sludge and garbage, to stimulate microbial activity.

The soluble salt content can be gradually leached down into the processed shale by the application of water, thus taking advantage of the limited permeability of the surface after treatment. This procedure also lowers the pH of the processed shale to a range which is normally encountered in soils in the Parachute Creek Basin.

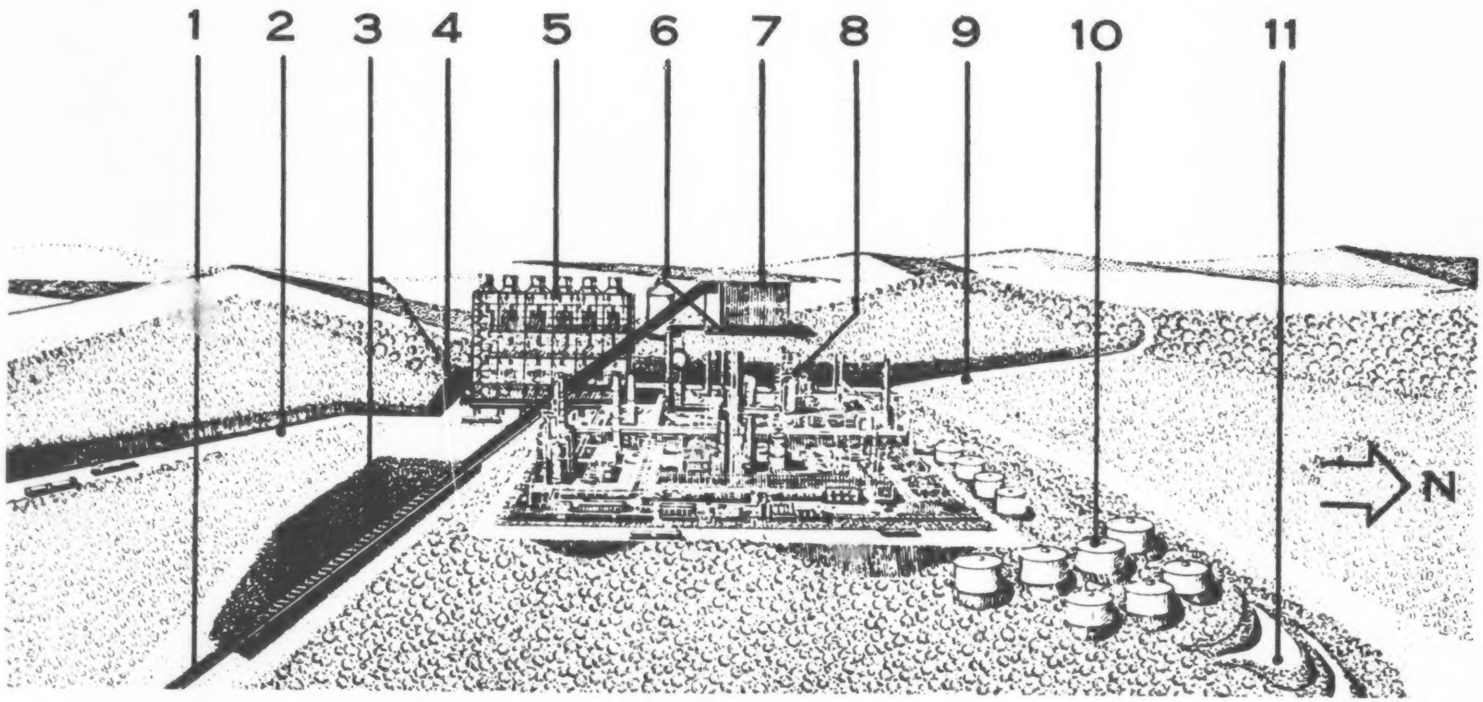
The structural characteristics of the processed shale can be improved to allow infiltration of water by addition of organic materials which would, as indicated, lower the pH, lighten the color, and raise the level of available nutrients. Such things as sawdust, peat moss, sewage sludge, manure, and ground garbage are being considered for this application. All of these would stimulate the growth of soil bacteria.

The dark color of the surface of the processed shale absorbs heat, and temperatures as high as 140° to 150°F have been observed in the top one-half inch of the compacted processed shale. This high temperature would, of course, inhibit germination. The addition



1 PLANT ACCESS ROAD  2 LOWER SEGMENT OF DAVIS GULCH  3 CATCHMENT DAM  4 MIDDLE FORK OF PARACHUTE CREEK
5 PROCESSED SHALE DISPOSAL IN DAVIS GULCH  6 REVEGETATED PROCESSED SHALE BENCHED EMBANKMENT  7 MINE BENCH
8 COARSE ORE CONVEYOR THROUGH TUNNEL FROM MINE BENCH  9 COARSE ORE CONVEYOR FROM TUNNEL TO FINAL CRUSHING
10 PROCESSED SHALE CONVEYOR  11 RETORTING & UPGRADING UNITS - PLATEAU SITE

FIGURE 5. SHALE OIL COMPLEX – COMPOSITE AERIAL VIEW



*Legend:*

1 COARSE ORE CONVEYOR 2 PLANT ACCESS ROAD 3 COARSE ORE STORAGE 4 PROCESSED SHALE CONVEYOR 5 PYROLYSIS UNIT 6 FINE ORE SILO
7 FINAL CRUSHING UNIT 8 UPGRADING UNITS 9 ROAD TO PROCESSED SHALE DISPOSAL 10 STORAGE TANKS 11 CATCHMENT BASINS

Figure 6. Retorting and Upgrading Units – Plateau Site

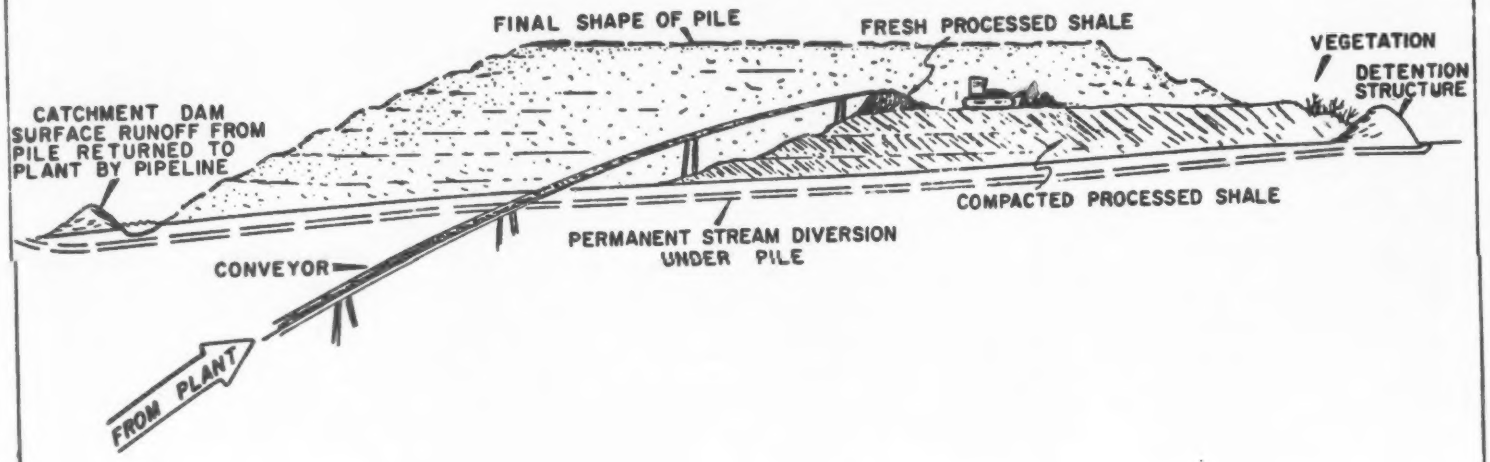

FINAL SHAPE OF PILE
FRESH PROCESSED SHALE
VEGETATION
DETENTION STRUCTURE
CATCHMENT DAM SURFACE RUNOFF FROM PILE RETURNED TO PLANT BY PIPELINE
COMPACTED PROCESSED SHALE
PERMANENT STREAM DIVERSION UNDER PILE
CONVEYOR
FROM PLANT

Figure 7. Cross Section of Processed Shale Disposal in Shallow Canyon or Valley (in 8" width)

of straw has been found to be most suitable in lightening the color and in helping to hold moisture near the surface, thus aiding germination. The addition of straw also reduces evaporation and aids the mechanical stabilization of the surface. And, of course, it eventually produces decayed organic material.

Investigations are under way to determine the suitability of placing a 6-inch cover of native soil on the processed shale embankment. This would be expected to provide for gradual water infiltration into the processed shale, resulting in an even more complete reduction of soluble salts. Revegetation would be more straightforward than with the processed shale itself.

The revegetation process will involve seeding with various grass and forb species, followed by watering and fertilizing. Plans are to fence out livestock and natural herbivores during the initial establishment phases. Even though investigations are still under way to determine the best species mixture and intensity of seeding, the following selection has been proposed by Dr. William Berg of Colorado State University:

| Common Name of Vegetation | Seeding Intensity (lb/acre) |
|---|---|
| Western Wheatgrass | 10 |
| Sodar Streambank Wheatgrass | 10 |
| Crested Wheatgrass | 6 |
| Indian Ricegrass | 2 |
| Sand Dropseed | 2 |
| Fourwing Saltbush | 9 |
| Russian Wildrye | 6 |

All species listed above are Colorado natives found in the Parachute Creek area.

The final goal of the processed shale disposal and revegetation is the formation of a plant community similar to that which naturally occurs in the Davis Gulch area. After leaching is completed and a sound grass cover is obtained, there are plans to plant a 2- to 4-year-old stock of native shrubs including Gambel oak, mountain mahogany, snowberry, serviceberry, and sagebrush. The ultimate combination of grasses and shrubs will blend in with the surrounding existing vegetation.

In the first 10 years of operation, there will be a need to revegetate about 9 to 14 acres per year. After 10 years of operation, there will be a need to vegetate over 100 acres per year. Using 1973 cost figures, the revegetation expenses will run about $1,600 per acre. This cost may be as high as $2,500 per acre if a 6-inch soil cover is applied.

## WATER QUALITY AND CONSUMPTION

The commercial plant planned by the Colony Development Operation will be located on the mesa top west of Middle Fork of Parachute Creek. A map of the Parachute Creek drainage system is shown in figure 8. The quality of water at the various sampling stations, shown in figure 8, illustrates the increase in salinity from upper reaches of the drainage to the point below Grand Valley. Sampling at Middle Fork (Station No. 551) gives a total dissolved solids of 461 parts per million, and at Davis Gulch (Sampling Station No. 552) the value is 446 parts per million. At Parachute Creek below Grand Valley, the total dissolved solids has increased to 791 parts per million. This increase in total dissolved solids is believed to be due to continual erosion of the soil and to the percolation of water through the underlying soil resulting in additional leaching of soluble salts. The net result is that Parachute Creek, below Grand Valley has a higher salt content than that of the Colorado River. This information illustrates the tendency toward increased salinity in this stream in the absence of industrial activity.

The Colony shale oil complex, designed to process 66,000 tons per day of oil shale, will require 3 to 4 barrels of water per barrel of produced oil. More than half of this water requirement will be used for moisturizing processed shale and for controlling dust, and will either become a permanent part of the processed shale or will be evaporated. This part of the water requirement can be obtained from impure water sources, such as highly saline aquifers and processed water from the oil shale complex. The remainder of the water requirement, needed for the production of steam, must be of higher quality and will be discharged into the atmosphere as water vapor. The upshot of this is that the processing complex will not require discharge of any water except as vapor. Thus, water use will be totally consumptive.

As previously discussed, the processed shale storage facility will be protected by a catchment dam designed to retain runoff from the maximum probable 1-hour thunderstorm. Therefore, almost all runoff from the surface of the processed shale embankment will be contained and reused. With no water discharge occurring from Davis Gulch, the Colony plant can then be considered a "zero discharge facility."

If under highly abnormal precipitation circumstances there is a discharge from the Davis Gulch catchment dam and spillway, it would be

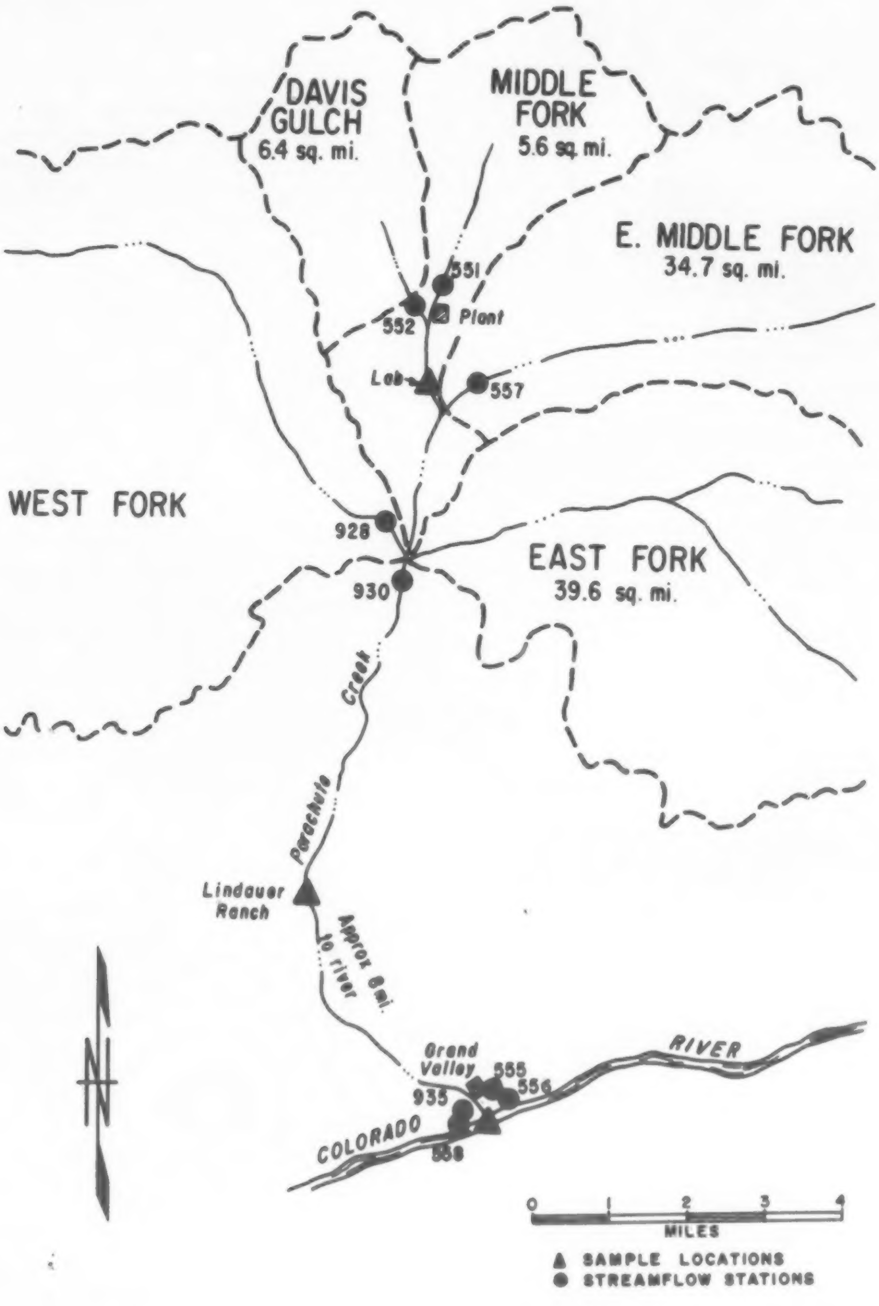

DAVIS GULCH
6.4 sq. mi.
MIDDLE FORK
5.6 sq. mi.
E. MIDDLE FORK
34.7 sq. mi.
551
552
Plant
Lab
557
WEST FORK
928
930
EAST FORK
39.6 sq. mi.
Parachute Creek
Lindauer Ranch
Approx 8 mi. to river
Grand Valley
555
556
935
558
COLORADO RIVER
N
0 1 2 3 4
MILES
▲ SAMPLE LOCATIONS
● STREAMFLOW STATIONS

Figure 8. Location of Stream Flow and Water Quality Stations (from Skogerboe 1973)

expected that the effect on base water quality would be insignificant for the following reasons:

1. Any precipitation associated with flood conditions would run off of the processed shale quite rapidly and would have minimum contact with the salt content of the processed shale, and thus not produce an abnormally high total dissolved solids content. During these periods of extremely heavy precipitation, any leachate from the embankment would be diluted with large volumes of natural runoff from other areas in Davis Gulch.

2. Conditions requiring spillway discharge from Davis Gulch dam will probably generate flood conditions in all portions of the Parachute Creek Basin, and these catchment dam discharges would be insignificant in their effect on the aquatic ecosystem.

3. It is generally agreed that long-term exposures to abnormally high concentrations of total dissolved solids and other pollutants would be required before plants and animals would begin to show observable adverse effects. Spillway discharges, if they occur at all, will not last for more than a brief period of time.

## SUMMARY

The Colony project on Parachute Creek is entering the plant construction phase with proven process and mining technology. Preservation of the environment has been a prime consideration in the development stages of this project and will contine to receive major attention during commercialization.

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15 May 1974

## Session IV:

## FUEL UTILIZATION AND TOTAL ENVIRONMENTAL ASSESSMENT

Paul Spaite
Session Chairman

# OVERALL ENVIRONMENTAL CONSIDERATIONS OF CONVERSION TECHNOLOGY

**C. E. Jahnig, E. M. Magee,
and C. D. Kalfadelis***

*Abstract*

*In view of the energy shortage, major efforts are underway to make better use of our coal and shale resources and to apply conversion technology to upgrade fuels. This paper discusses environmental considerations that result from the application of conversion technology, describes the work underway in this area, and gives some of the results to date. Specific examples include:*

1. *Cleanup and waste heat recovery on the raw gas from gasification.*
2. *Considerations on ash disposal, leachability, and trace metals.*
3. *Modifications in coal preparation and drying to improve fuel efficiency, dust recovery, and control of sulfur in the vent gas.*
4. *The need for a coal-fired utility boiler and alternatives available to control emissions from it.*

*Objectives of the studies will be discussed as well as the approach and methods used.*

## INTRODUCTION

A major effort is underway to make better use of our coal and shale resources in order to alleviate the energy shortage. This work necessarily involves conversion technology to upgrade these solid fuels to gas or liquid fuels which will cause less pollution and which will be easier to use. Environmental considerations of the conversion process are discussed below, together with some of the background on objectives and approaches used in analyzing environmental aspects of various processes.

A primary objective of this work, which is being carried out under contract with the Environmental Protection Agency,† is to recognize and point out potential problems before the processes reach a commercial stage, so that practical solutions can be developed in a well-planned approach, rather than waiting for the problem to reach a stage of urgency. In analyzing various conversion processes, it also becomes apparent that some areas are not yet adequately defined and that further information, and perhaps additional experimental work, is needed to provide a suitable basis for evaluation. Some of the problems are common to more than one conversion process, such as coal drying. In such cases, a coordinated program can work out the solutions for general use instead of each developer having to do all of the work himself. One desirable objective, therefore, is to define common problems that can benefit from a coordinated effort.

## ENVIRONMENTAL CONSIDERATIONS

Aspects of a process that may affect the environment are listed in table 1. Each of these must be examined carefully for every specific case. Consideration of effluents to the air will include sulfur and nitrogent oxides, hydrocarbons, odors from phenols and ammonia, particulates, etc. Water effluents may contain sulfur compounds, ammonia, cyanides, phenols, oil, particulates, etc. Cleaning up water for reuse so that there is no wastewater effluent from the plant except for that used to slurry the ash being disposed of is desirable and is generally practical as well.

The ash may be returned to the mine or used for landfill, which raises the question concerning leachables such as calcium chloride, magnesium sulfate, fluorides, etc. It is apparent that further information is needed in this area.

Table 1. Environmental considerations

| | |
|---|---|
| Air effluents | Thermal effluent |
| Water effluents | Odor and taste |
| Ash disposal | Noise |
| Trace elements | Visual |
| Water consumption | Land use |

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*The authors are in the Government Research Laboratory of Exxon Research and Engineering Company, Linden, New Jersey.

†This work was carried out under Contract No. 68-02-0629 with the Environmental Protection Agency.

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Trace elements are also part of this consideration (ref. 1). Most of the heavy metals in the coal feed remain with the ash during gasification or liquefaction. However, some of the trace elements volatilize to a moderate or large extent during gasification. These elements probably do not remain in the product gas but rather are rejected in some other effluent stream. Therefore, we need to find out about the composition and leachability to see if corrective measures are needed. Of the various elements so far designated as toxic, it is known that all of them volatilize to a moderate or large extent during gasification. They may then build up in the recirculated water used to scrub the raw gas or possibly interfere with the amine or other system used to remove sulfur.

Water consumption is a growing concern, especially at Western locations. The major consumption is usually from evaporation in the cooling tower. Reducing the cooling water requirement will therefore cut the makeup water consumption. Application of air-fin cooling can help, together with heat exchange between streams to increase thermal efficiency.

Thermal efficiency is important in that essentially all of the unrecovered heat is taken up by air or water in the environment. It also reflects how much raw material is consumed in making a given amount of clean fuel.

## OBJECTIVES AND APPROACH

In analyzing a process from the environmental standpoint, the objectives and approach that we have used are shown in table 2. First, all streams entering and leaving the plant are carefully defined, insofar as possible, as to amount and composition. For many of the streams, exact information is not available and reasonable assumptions have to be made. Also, the entire heat balance is worked out thoroughly to determine the overall thermal efficiency of the process, to see where the major losses are, and to determine where potential improvement may be possible. The major pollution problems are then pinpointed. The next step is to see to what extent these can be alleviated or eliminated by simple engineering-type modifications—for example, recycling wastewater rather than discharging it to a river. Where the problem cannot be avoided, additional control facilities are added, as in the case of coal drying where bag filters, a scrubber, or electrostatic precipitator may be needed to control dust.

Table 2. Objectives

(1) Define all streams in and out--amount, composition, and energy balance.

(2) Add pollution control facilities as needed, indicate alternatives.

(3) Point out simple modifications and potential improvements to decrease emissions or increase efficiency.

(4) Identify areas where specific additional information is needed well before commercial use.

(5) Determine technology needs for pollution abatement.

One specific objective of analysis is to point out where further work is necessary or desirable to resolve environmental questions, or where significant improvements in the operation may be possible. Some of these will be described later in this paper, and in a subsequent paper (ref. 2). Of particular interest are areas that are common to more than one process, such as coal preparation, storage, drying, and grinding.

The system for cleaning up the raw gas from coal gasification has common features for most of the processes as outlined in table 3. High-energy water scrubbing is generally needed to remove dust. This necessarily means that the gas has been cooled to the water dewpoint. It is important to recover as much of this heat as possible in order to minimize the amount of heat taken out by air-fins or cooling water. The scrubber water will pick up large amounts of hydrogen sulfide and ammonia. Techniques are available for sour water stripping in order to produce separate streams of hydrogen sulfide and ammonia that are relatively pure. Ammonia may constitute a byproduct for sale while hydrogen sulfide can go to the sulfur recovery plant.

Some processes also produce significant amounts of tar, naphtha, cresols, phenol, etc., in which case they will have to be recovered effectively. They might be sold, burned as fuel, or recycled to the conversion process. Cleanup of the water layer will involve most of the steps indicated on the slide, depending on what

Table 3. Typical raw gas cleanup

Cooling: Steam, C.W., Air-fins

Dust Removal: Scrub, Bags, Elect. Precip.

Tar, Oil, Naphtha

Phenols, Cresols, etc.

$NH_3$, CN, SCN, Compounds

$H_2S$, COS, Mercaptan, Thiophene

Other: Carbonyls, Arsine, Fluoride

Water Layer: Strip, Extraction, Biox, Filter, Activated Carbon, Sludge Incin. (Chemical Additions).

materials are present and on how the water is disposed of.

## EMISSIONS CONTROL

A single sulfur removal system that would take out all forms of sulfur present in the raw gas would be desirable, but such a system is not available today. Characteristics of sulfur removal techniques are shown in table 4. $H_2S$ is readily removed, but not carbonyl sulfide, carbon disulfide, or thiophene. Moreover, the usual amine scrubbing systems take out much of the carbon dioxide present along with $H_2S$. Unfortunately, this gives a low concentration of $H_2S$ going to the sulfur recovery plant; where a Claus Plant is used, it results in lower sulfur recovery and higher cost. Hot carbonate will remove much of the carbonyl sulfide but is limited in that it also takes out much of the carbon dioxide, resulting in a low concentration of $H_2S$ going to the sulfur recovery plant.

Various processes are available for cleaning up tail gas from a Claus unit including the Beavon, Clean-Air, SCOT, IFP, and Wellman-Lord (ref. 3). The first three of these depend on reducing sulfur compounds to $H_2S$ which is then removed. IFP uses a liquid phase Claus-type reaction but does not remove carbonyl sulfide or carbon disulfide. Wellman-Lord uses a sulfite solution to scrub out $SO_2$, thus making sulfuric acid as a byproduct.

Consideration should also be given in gas cleanup to using one of the absorption/oxidation type of processes as offered by Stretford, IFP, Tackahax, and others. These make sulfur directly and can reduce $H_2S$ to a very low level even in the presence of considerable $CO_2$, but they do not remove carbonyl sulfide.

Many gasification processes generate other chemical compounds such as phenol, cyanides, and tars. Although these should be removed in the gas cleanup section, some may remain and interfere with the operation of the other parts of the process. For example, cyanides interfere with the Stretford-type sulfur removal, while in the case of amines, thiocyanates will accumulate and have to be purged.

There is a universal need for plant utilities. Generally, a power plant is included to make high pressure steam either for the process or for generating electric power. Using purchased power does not avoid this question but only transfers it to a different area. Utilities are a large fuel consumer. They often consume 15 to 25 percent of the total coal used by a process. If the fuel fired to this utility boiler is high-quality product gas, then it reduces thermal efficiency and is expensive. However, gas may be used to the extent needed to control sulfur emission. One route is to make low-Btu fuel gas in a separate gasification system using air instead of oxygen. An alternative to consider carefully is burning coal or char in this utility boiler and adding stack gas cleanup to control sulfur and dust emission. For those processes which necessarily produce byproduct char, this may be the only alternative to gasifying the char. A number of commercial processes are offered for stack gas cleanup and it becomes a choice for the particular application as to what system is actually used.

Comparisons are needed between the emission

Table 4. Sulfur recovery

MEA - High $H_2S$ capacity, also removes $CO_2$, not COS.

Higher Amines - More selective $H_2S$ vs. $CO_2$ but low capac.

Carbonates - Remove COS but low selective $H_2S$ vs. $CO_2$.

Claus Plant - Inefficient if less than 20-25% $H_2S$ in feed.

Absorp./Oxid. - Selective for $H_2S$, not good on COS.

Sulfur Guard - Need reheat to 600°F.

Table 5. Emission standards

| Fuel | lb/MM Btu | | |
|---|---|---|---|
| | Dust | $SO_2$ | $NO_2$ |
| Gas | 0.1 | -- | 0.2 |
| Liquid | 0.1 | 0.8 | 0.3 |
| Solid | 0.1 | 1.2 | 0.7 |

Water:

Current - all cyanides, compounds of Hg. Cd.

Potential - As, Se, Cr, Zn, Pb, Be, Ni, Sb.

standards and actual plant effluents, many of which are not completely defined at this time. Some of the standards are shown in table 5. The upper part applies to large stationary power plants firing various fuels. The lower part of the table indicates some of the water contaminants that now have limits and others for which emission standards may be expected in the future.

## ALTERNATIVES TO CONSIDER

Examples of some engineering-type modifications and improvements will now be given. Some of these examples are shown in table 6.

One alternative is in the type of coal-drying. It is common to operate a coal dryer with a large amount of excess air such that the oxygen level is around 10 to 11 percent. This makes for efficient drying in that the gas volume is very large so that the evaporated moisture contributes less to the humidity level. However, it also means that a large volume of gas must be handled and cleaned up. Serious consideration should be given to reducing the gas volume as much as possible and allowing its moisture content to increase. While this will tend to make the drying less efficient, it may not be an overriding factor in that the coal will eventually be heated to much higher temperatures which will drive off any residual moisture. In fact, the trend appears to be to preheating the coal to higher temperatures, such as 500°F, so as to reduce the heat load in gasification or liquefaction, but not to drive off volatiles in the drying or preheating operation. In effect, what is proposed is simply to run the coal dryer firing minimum practical excess air. This will reduce the gas volume and also increase the fuel efficiency. Whereas a typical fuel efficiency in drying may be 60 percent, it then becomes possible to achieve 80 percent fuel efficiency. This is no small item since the amount of fuel used in the coal preparation may be 10 percent of the total coal input. For the operation at low excess air, it may become very desirable to recover water from the dryer vent gas in order to reduce the overall water consumption of the process. Again, this is a significant item in that with western lignite of say 33 percent moisture content the water recovered could cut in half the overall makeup water required.

Table 6.

Examples of alternatives to be considered:

(1) Type of coal-drying

(2) Type of fuel to coal dryer

(3) Hydrogen production

Another place where alternatives must be considered as in the fuel to be fired in the coal dryer. This could be high quality gas to give minimum sulfur emission. It is more efficient to burn coal in the dryer, and where this is done it can add about 1 percent to thermal efficiency. Some processes cannot use the fines produced in grinding so a convenient place to use them efficiently is in the coal dryer. For sulfur control, part of the fuel may be low sulfur, low-Btu gas drawn off before methanation. Vent gas from the dryer must be cleaned up to remove dust or coal fines, so it is possible that sulfur control might be included in the dust removal system, particularly if it involves scrubbing. One possibility is the addition of limestone to the scrubber water. This of course will depend on the amount of dust reaching the scrubber, its combustible content, and whether it needs to be reused.

In many conversion processes, hydrogen manufacture is required to supply hydrogen for coal conversion or for product treating. The hydrogen might be made by gasification of char or heavy liquids. In the case of coal liquefaction there may be sufficient byproduct hydrocarbon gas to provide hydrogen by conventional steam reforming of the gas. In any event these operations must be included in the overall balances.

As has been pointed out, the utilities system and coal preparation are both major factors in the environmental aspects of a process. They are common to a large number of conversion processes and it is desirable to define simple alternative systems that can use coal as fuel and still provide adequate environmental controls.

## STATUS OF WORK

The status of our work at the present time is that detailed analyses of a number of coal gasification processes have been made. One of these has been published (ref. 4). Others will be published shortly. Several other analyses are underway, including retorting and liquefaction processes. Other studies will be made as needed, for example, on retorting oil shale and on liquid hydrocarbon gasification. These will provide a framework to use in accessing the environmental impact of various conversion processes to upgrade fossil fuels.

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4. E. M. Magee, C. E. Jahnig, and H. Shaw, "Evaluation of Pollution Control in Fossil Fuel Conversion Processes - Gasification, Section 1: Koppers-Totzek Process," EPA Report No. EPA-650/2-74-009a, January 1974.

# WEIGHING ENVIRONMENTAL COSTS AND BENEFITS

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*Abstract*

*A methodology has been developed to assess the environmental factors related to a selected number of fuel/energy systems. This methodology involves the compilation and organization of effluent data, the evaluation of the combined effects of extraction, transportation, processing, and utilization of fuel to produce energy. It also provides a technique for ranking the fuel/energy systems from an environmental standpoint. The principal objective in the development of the methodology was to provide a basis for making judgments regarding economic and environmental tradeoffs.*

*The utilization of this methodology has led to the conclusion that air emissions associated with coal utilization can be decreased to approximately the equivalent of those being emitted from systems using natural gas. However, there is an attendant increase in the environmental burdens imposed on the water and land media. This improvement can be accomplished through the use of the advanced control technology expected to be available during the early part of the 1975-1990 time period. Later in that period, additional technological advances will lead to procedures and systems for minimizing the burdens imposed on the water and land media by the utilization of coal.*

## INTRODUCTION

The desire to obtain the benefits of pollution control is reflected in the establishment of laws, programs, and policies designed to improve the quality of the environment by controlling emissions at the expense of economic impacts upon society. It is not easy to estimate the benefits which may derive from these expenditures, although estimates based upon the data available and on certain assumptions are available.

The fact that certain benefits have not been quantified or valued in economic terms does not mean that these benefits are unimportant. On the contrary, this importance is revealed by the fact that most of the goals usually stated tend to deal with benefits that are typically not measured. All benefits (both measured and unmeasured) must be considered in any decision, when analysis involves comparing the costs of proposed actions with desired or expected benefits.

A formal procedure for assessing the tradeoff between benefits to society and costs does not exist. Currently such tradeoff analysis is done on a judgment basis, often in the political arena.

The benefits of pollution abatement are obtained in two ways. One is to reduce the existing pollution level to some target level by controlling emissions to reduce the level of pollution costs. The second way is to prevent pollution levels from becoming worse in order to avoid incurring additional pollution costs. In evaluating the effectiveness of current or proposed programs, the abatement costs should be compared to the sum of the reduced pollution costs and the avoided pollution costs.

In order to understand better the benefits of pollution control, it may help to distinguish among three types of costs: (1) damage costs, (2) psychic costs, and (3) avoidance costs. The psychic costs imposed by pollution are distinguished from damage and avoidance costs in that no out-of-pocket expenses are involved; people simply tolerate:

(1) the mental discomfort or anguish persons feel because they perceive a threat in pollution becoming worse;
(2) the discomfort resulting from direct exposure to the pollutants like smarting eyes, shortness of breath, and physical weakness;
(3) the loss in pleasure because there is reduced sunlight, restricted visibility, increased discoloration of buildings, and damaged or discolored vegetation;
(4) the mental discomfort or anguish that some persons feel because they believe nature is being assaulted and the esthetic quality of life is being degraded.

When empirical estimates of pollution costs are made, it is not always possible to identify which categories of pollution costs are being measured. For example, property value estimates may include some of all three kinds of pollution costs.

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### *Methods of Assessing Pollution Costs*

What are the methods that can be used to measure society's willingness to pay for improved air quality? There are six basic methods that can be used: (1) valuing physical (dose-response) relationships; (2) market studies; (3) opinion surveys of air pollution sufferers; (4) litigation surveys; (5) political expressions of social choice, (6) the delphi method. Each method has been used under different circumstances with varying degrees of success.

The most widely used technique is that of determining a physical (dose-response) relationship between a pollutant and an object or living thing. These relationships are determined either by designed experiments or by analysis of many observations of natural events. The physical relationship is then transformed into economic terms by determining values for the effects. The aggregate or national damage estimate is obtained by determining the population exposed to various levels of the pollutant.

In the market study approach, pollution damages are judged on the basis of human behavior as reflected in specified markets. This approach completely circumvents the need to know the physical or biological damage function. If people are willing to pay to avoid the effects of pollution, then property values and local environmental quality will vary inversely. A significant problem in using the market study approach is that all the factors that explain consumer preferences and behavior must be included in the analysis.

The use of the opinion survey is closest to the classical economic approach in that it focuses on estimating utility and demand functions. Investigators employing this method have attempted to ascertain what people do and do not perceive as pollution effect. Opinion surveys have shown particular usefulness in understanding how attitudes about air pollution are formed and then affected by changes in air quality.

Litigation surveys could be used to determine the extent to which people have turned to the courts for redress for pollution damages; the use of this method is limited.

An investigator may try to gauge political expressions, representations, and exhortations in the hope that their intensity somehow corresponds to the intensity of preference for one outcome over another.

In the delphi method, the knowledge and abilities of a diverse group of experts are pooled for the task of quantifying variables which are either intangible or shrouded in uncertainty. The use of this method provides an efficient way to obtain best judgments from the knowledge and opinions of experts, even though these judgments and decisions may be essentially subjective. This approach may be broadened to include actual technical estimates.

Of these methods, valuing dose-response relationships, and a particular market study application—called the property value method—have yielded the most promising insights into the true nature of air pollution damages. With effective abatement, these damages become the benefits of control. Yet, even the application of these methods has been fraught with many problems. It is difficult to allocate the observed damages among a number of synergistically interacting multiple stresses to the environment. The damages themselves cannot be easily measured and reduced to economic terms.

### *An Example: Air Pollution Cost Estimates*

The benefit to be expected from controlling air pollutant emissions to meet the established or assumed ambient air quality standard should only be compared to the increment of abatement costs incurred to reduce pollution to this level. In table 1 are presented estimates of the reduction in some of the pollution costs that could result from reducing the 1970 level of certain air pollutants to meet the current standards. Not all of the costs have been estimated; for example, the damages to animals and the natural environment have not been obtained. This does not imply that such pollution costs do not exist, but only that there is not enough information to make an estimate. The wide range of these estimates implies that little confidence can be placed on the best estimate.

The estimate of esthetic and soiling costs was obtained from a study of property values which provided a measure of the psychic costs, damage costs, and avoidance (including adjustment) costs that people suffer because of sulfur oxides and particulates. This value was obtained from original study values by adjustment to avoid the double counting of health and materials damage effects.

The estimates for health costs measure the value of damages resulting from air pollution effects--reduced productivity because of ill health or premature death, and out-of-pocket health care expenses. Data concerning the effects of oxidants (primarily hydrocarbons and oxides of nitrogen) and of carbon monoxide did not allow for the estimation of the value of damages by these pollutants. Psychic and avoidance costs are also omitted from these health estimates.

The materials estimates measure the value of

Table 1. National Estimates of Air Pollution Costs, by Pollutant and Effect, 1970
($ billion)

| Effects | Sulfur Oxides | | | Particulates | | | Oxidants[a] | | | Carbon Monoxide | Total | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Low | High | Best | Low | High | Best | Low | High | Best | Best | Low | High | Best |
| Aesthetics & Soiling[b,c] | 1.7 | 4.1 | 2.9 | 1.7 | 4.1 | 2.9 | ? | ? | ? | * | 3.4 | 8.4 | 5.8 |
| Human Health | 0.7 | 3.1 | 1.9 | 0.9 | 4.5 | 2.7 | ? | ? | ? | ? | 1.6 | 7.6 | 4.6 |
| Materials[c] | 0.4 | 0.8 | 0.6 | 0.1 | 0.3 | 0.2 | 0.5 | 1.3 | 0.9 | * | 1.0 | 2.4 | 1.7 |
| Vegetation | * | * | * | * | * | * | 0.1 | 0.3 | 0.2 | * | 0.1 | 0.3 | 0.2 |
| Animals | ? | ? | ? | ? | ? | ? | ? | ? | ? | * | ? | ? | ? |
| Natural Environment | ? | ? | ? | ? | ? | ? | ? | ? | ? | ? | ? | ? | ? |
| Total | 2.8 | 8.0 | 5.4 | 2.7 | 8.9 | 5.8 | 0.6 | 1.6 | 1.1 | ? | 6.1 | 18.5 | 12.3 |

[a] Also measures losses attributable to oxides of nitrogen

[b] Property value estimator

[c] Adjusted to minimize double-counting

? Unknown

* Probably negligible

Sources: Waddell, Thomas E., "The Economic Damages of Air Pollution: Unpublished Report, EPA National Environmental Research Center, Research Triangle Park, March, 1974.

damages and some of the avoidance costs resulting from air pollution damage to manmade materials. It is impossible to say what portion of avoidance costs are accounted for. Estimates of the value of air pollution effects on plants mostly represent the direct damages and generally ignore the avoidance costs and psychic costs.

The damage to animals caused by air pollution has generally been localized, and its economic consequences have probably been relatively unimportant. Little is known about the effects of air pollution on domestic animals and wildlife. Also, little is known about how these problems interface with the natural environment.

The natural environment category includes the pollution costs of: disruption or destruction of ecological systems, the destruction of species, the disruption of social systems or social patterns, and the disruption of a man-nature balance. Many of these pollution costs affect man as psychic cost, particularly fear of man's inadvertently destroying life on earth. Because they are mostly psychic costs, there is great difficulty in quantifying them, and no estimates are yet available.

### *The Cost/Benefit Comparison*

It is desirable for any control program, policy, or action, that the benefits of reduced pollution costs be greater than the abatement costs; otherwise society will be made worse off. In fact, it is very difficult to obtain accurate enough cost and benefit values for a given program, policy, or action to make an accurate determination of the value of the action to society. The available data do not provide an adequate basis for accurate comparison of the cost and the benefit. A comparison of national costs and benefits would not be sufficient to judge the merit of pollution control programs for individual regions of the country or for individual pollutants.

## FOSSIL-FUEL ENERGY SYSTEMS

The attempts to assess the costs and benefits of pollution control and the difficulties encountered have been described in the preceding sections of this paper. An alternate and perhaps simpler approach is to analyze the various systems which can be used in the generation of a product required by society and to attempt to determine which system imposes the least burden on the environmental media. An example of such an analysis is a study which Battelle-Columbus conducted for the Environmental Protection Agency a little more than a year ago. This was done in order to provide a basis for recommending environmentally preferred energy policy initiatives which may be required to meet future energy demands. The overall objective of Battelle's efforts was to provide EPA with the necessary information regarding the environmental emissions of alternative energy cycles so as to permit explicit judgments regarding economic and environmental tradeoffs. This will permit the formulation and execution of sound policy in this critical area.

The first task was concerned with a projection of the environmental burden of baseline fuel-supply projections during the period of 1975 to 1990. This effort was based on a comparative analysis of published long-term fuel supply and demand projections. This was accomplished for both existing and developmental technologies involved in the various segments of the energy systems. Task 1 thus included the identification and quantification of the environmental emissions of alternative fuel supplies commercially available in the 1975 to 1990 time period. The sources of energy under consideration in this portion of the study included coal, oil, natural gas, and nuclear fission. Because of the short time available for this study, the impacts were quantified in terms of effluent quantities but not in terms of effluent effects. It further was specified that the utilization segment of the energy cycle would be concerned only with the generation of electrical power and with space heating.

The second task included a quantification to the maximum extent possible of the effectiveness and economic costs of pollution control for each alternative energy supply and technology considered in Task 1. These were considered in terms of common indices to permit comparison of alternatives. Wherever possible, pollution control was considered as that necessary to achieve existing standards or new standards which are anticipated as a result of Federal or environmental legislation. Alternatively, pollution control would apply the "best available" technology.

The third task was the ranking of all alternative energy supplies and technologies from best to worst, so far as environmental burden is concerned. The environmental effects of each phase of the energy cycle were considered, and separate rankings made for the commercially available energy supplies and for the development technologies.

For each of the tasks, the information analyzed by Battelle was drawn from published literature, from experience of Battelle staff members in various fields, and from members of the EPA staff.

The very broad scope of the subject and the short time available for the preliminary study limited the effort to an overview. A data base of environmental emissions was compiled from readily available information, and a preliminary methodology was developed for ranking the fuel/energy systems on the basis of environmental burden. These elements are extant and can serve as a framework for more detailed analysis and for the addition of new data on emissions and technologies as it is developed, if the program is extended and expanded.

For purposes of this symposium, only the fossil fuel segment of the electrical energy industry will be considered in detail. However, nuclear energy systems will be included in the overall ranking.

It is anticipated that in the time period of concern to the study—1972 to 1990— fossil fuels will continue to be the dominant source of energy in the United States. For this reason, considerable attention has been given to the analysis of the many and varied energy systems currently in use or under development which utilize fossil fuels.

The complexity of the environmental factors associated with the utilization of fossil fuels requires that the environmental impact of alternative energy sources must be analyzed for the entire energy system from extraction through utilization. This, in turn, requires a systematic approach which will accommodate the large number of variables involved and which will result in an evaluation of energy systems which reflects all of the variables. The modular approach chosen for this study permits such a systematic evaluation. Because of the limitations in the data base, the environmental impact of each module has been evaluated on the basis of emissions, but not in terms of effects, as ultimately should be done to permit development of a more meaningful ranking methodology.

### *Modular Approach*

The pathways by which various fossil fuels are utilized or processed and converted to other forms of energy are many and varied. in order to assess the efficiency of energy utilization and the environmental burden for the various optional energy systems which utilize fossil fuels, a modular approach has been used. In general, the modules, defined as a distinct phase in a fuel/energy system, fall within one of the five categories: the extraction or procurement of the fuel from its source, the processing of the fuel, the conversion of the fuel to a different form of energy, the transportation, or the utilization of the fuel. This approach allows ready identification of those phases of the fuel/energy system which contribute a major share of the environmental burden. It also provides a data base for extensive analysis of a large number of possible system options. A list of the important modules to be considered is given in table 2.

A very large number of possible pathways from the extraction of a fuel to utilization of the fuel exist. A conceptual view of the modular relationships of importance are presented for coal, oil, and gas in figures 1 through 5. From these diagrams, a list of systems composed of from two to five modules was constructed and is presented in table 3. The list does not represent all possible combinations of modules but rather those considered to be of major importance.

Fifty selected modules were analyzed for energy efficiency and environmental burden during the course of the first segment of the study. A list of these modules is given in table 4.

### *System Description and Assessment*

The modules described in the foregoing section were used as building blocks for the various potential energy systems. In the study of electric power generation systems, 15 selected coal systems, 2 mixed-fuel (coal and municipal refuse) systems, 6 oil systems, and 2 gas systems have been described and analyzed. These include the important options open to the Nation for meeting the projected energy deficit. In addition, 5 systems have been analyzed in which the end use of the energy is space heating. The overall environmental burden for each energy system has been obtained through the summation of the estimated environmental burden for each module making up the system, and modified by the efficiency of energy utilization for these modules. Weighting factors were applied to permit a summation of burdens.

In addition to the summation of environmental burdens, costs of energy conversion for each of the systems were estimated so that the systems could be compared economically, as well as environmentally. The cost of pollution control for specific modules has been developed for those modules for which this information is available.* In addition, the overall cost of energy production has been derived for each system.

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*Report referred to in "Acknowledgment" at end of this paper.

## Table 2. Modules of Significance in the Analysis of Environmental Impacts

### Gas Extraction, Processing, Transportation, and Utilization Options

EXTRACTION AND CLEANING

Gas Extraction and Cleaning/Continental (North American Exclusive of Arctic Regions)
Gas Extraction and Cleaning/Off-Shore
Gas Extraction and Cleaning/Arctic
Gas Extraction and Cleaning/Overseas

TRANSPORT

Gas Pipeline (Conventional)
Gas Pipeline (Arctic)
Cryogenic Tanker

STORAGE

Storage/Domestic
Storage/Tanker

UTILIZATION

Conventional Boiler
Combined Cycle
Space Heating

### Oil Extraction, Conversion, Secondary Processing, Transportation, and Utilization Options

EXTRACTION

Oil-Gas Well/Continental
Oil-Gas Well/Off-Shore
Oil-Gas Well/Overseas

TRANSPORT OF CRUDE

Oil Pipeline (Conventional)
Oil Pipeline (Arctic)
Tanker

CONVERSION OF CRUDE

U. S. Refinery
Topping Operations (Overseas)
Topping Operations (Domestic)

PROCESSING OF FRACTIONS

Residual Desulfurization
Naphtha No. 2 Fuel Production
Heavy Oil Gasification
Light Oil Gasification
Blending Fuel Oil

TRANSPORT OF PRODUCTS

Barge
Tanker
Pipeline

UTILIZATION

Conventional Boiler
Conventional Boiler/Flue Gas Cleaning
Combined Cycle
Fluid Bed Combustor
Space Heating

### Coal Extraction, Processing, Transportation, and Utilization Options

EXTRACTION

Surface Mine (Eastern Coal)
Surface Mine (Western Coal)
Underground Mine (Eastern Coal)

TRANSPORT OPTIONS (Before Processing)

Rail
Barge

TREATMENT OR CONVERSION PROCESSES

Physical Coal Cleaning
Chemical Leaching
Solvent Refining
Gasification (Low Btu)
Gasification (High Btu)

TRANSPORT OPTION (After Processing)

Rail
Barge
Coal Slurry Pipeline
Gas Pipeline

UTILIZATION PROCESSES

Conventional Boilers
Combined Cycle
Fluid Bed Combustor
Conventional Boiler/Flue Gas Cleaning
Space Heating

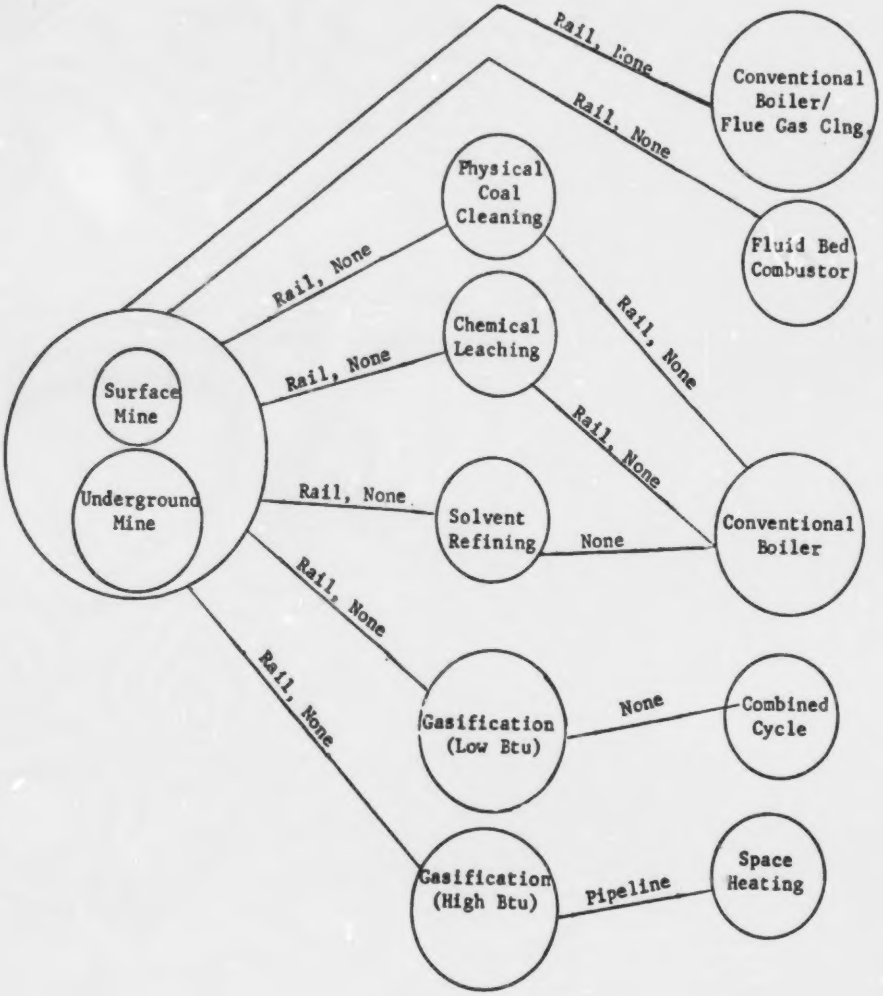
Rail, None
Conventional Boiler/ Flue Gas Clng.
Rail, None
Physical Coal Cleaning
Fluid Bed Combustor
Rail, None
Rail, None
Chemical Leaching
Rail, None
Surface Mine
Rail, None
Underground Mine
Rail, None
Solvent Refining
None
Conventional Boiler
Rail, None
Rail, None
Gasification (Low Btu)
None
Combined Cycle
Gasification (High Btu)
Pipeline
Space Heating

Figure 1. Schematic Representation of Modular Relationships-- High Sulfur Eastern Coal

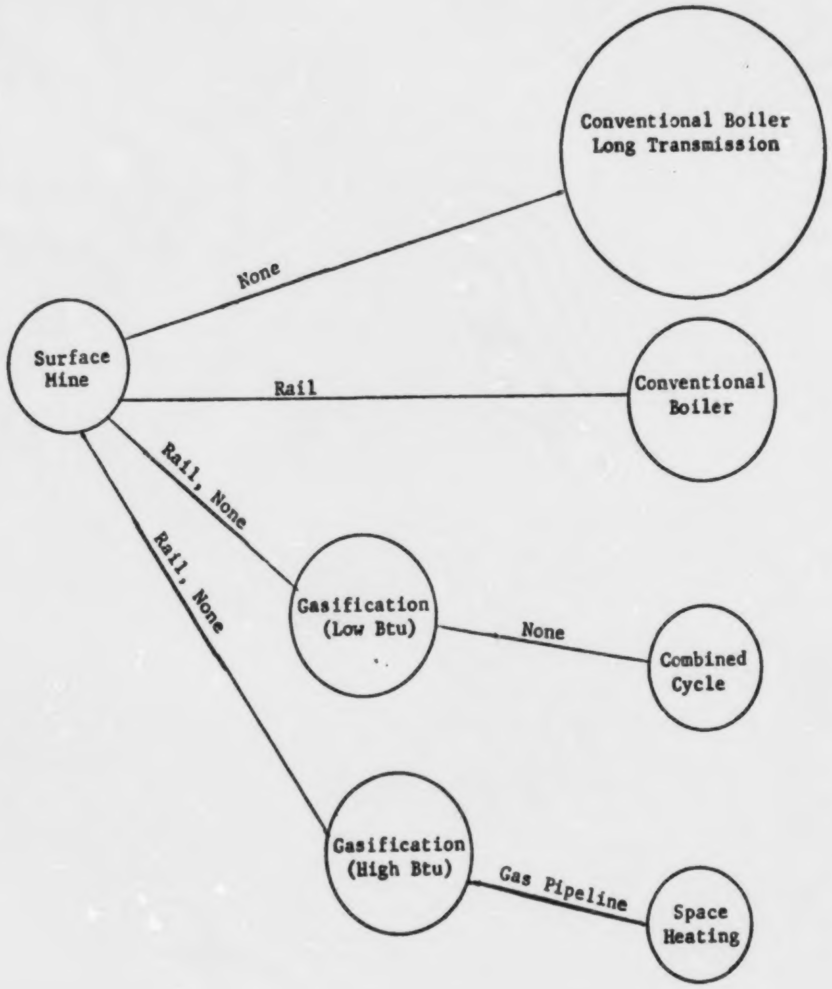
Conventional Boiler
Long Transmission
None
Surface
Mine
Rail
Conventional
Boiler
Rail, None
Rail, None
Gasification
(Low Btu)
None
Combined
Cycle
Gasification
(High Btu)
Gas Pipeline
Space
Heating

Figure 2. Schematic Representation of Modular Relationships -- Low Sulfur Western Coal

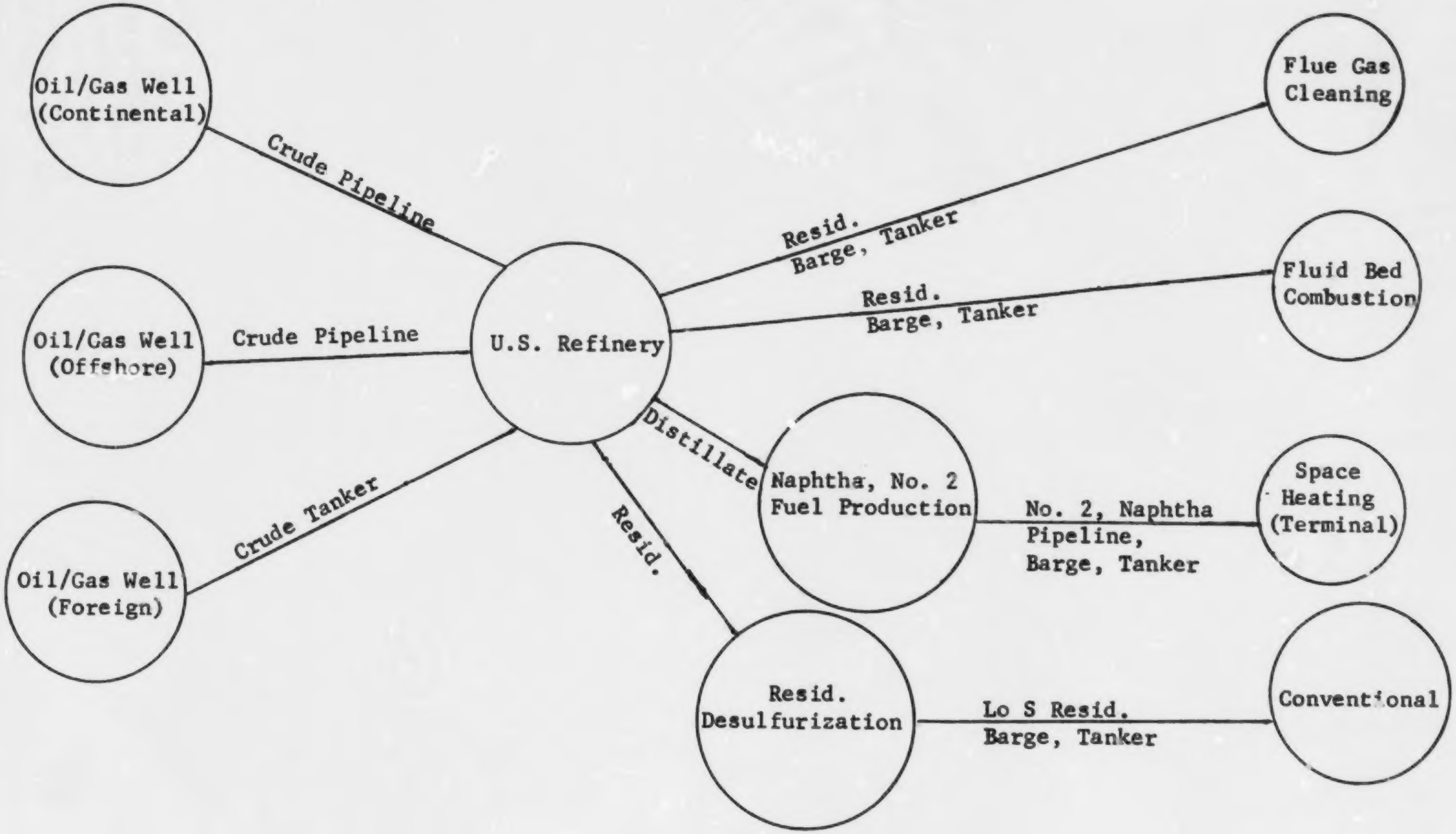
Oil/Gas Well (Continental)
Oil/Gas Well (Offshore)
Oil/Gas Well (Foreign)
Crude Pipeline
Crude Pipeline
Crude Tanker
U.S. Refinery
Resid. Barge, Tanker
Resid. Barge, Tanker
Distillate
Resid.
Flue Gas Cleaning
Fluid Bed Combustion
Naphtha, No. 2 Fuel Production
No. 2, Naphtha Pipeline, Barge, Tanker
Space Heating (Terminal)
Resid. Desulfurization
Lo S Resid. Barge, Tanker
Conventional

Figure 3. Schematic Representation of Modular Relationships -- Oil

Table 3. Summary of the More Important Systems Options

| Extraction Process | Transport | Processing of Raw Fuel | Secondary Processing | Transport & Storage | Utilization |
|---|---|---|---|---|---|
| | | **Fuel/Energy Systems - Coal** | | | |
| (1) Surface Mine<br>(2) (Eastern Coal) | None | Physical Coal Cleaning | None | Rail, Barge | Conventional Boiler |
| (3) Surface Mine<br>(Eastern Coal) | None | Chemical Leaching | None | Slurry Pipeline | Conventional Boiler |
| (4) Surface Mine<br>(Eastern Coal) | None | Solvent Refining | None | None | Conventional Boiler |
| (5) Surface Mine<br>(6) (Eastern Coal)<br>(7) | Rail, Barge, None | Gasify (Low Btu) | None | None | Conventional Boiler |
| (8) Surface Mine<br>(9) (Eastern Coal)<br>(10) | Rail, Barge, None | Gasify (Low Btu) | None | None | Combined Cycle |
| (11) Surface Mine<br>(12) (Eastern Coal)<br>(13) | Rail, Barge, None | Gasify (High Btu) | None | Pipeline Gas | Space Heating |
| (14) Surface Mine<br>(15) (Eastern Coal)<br>(16) | Rail, Barge, None | None | None | None | Conv. Boiler with Flue Gas Cleaning |
| (17) Surface Mine<br>(18) (Eastern Coal)<br>(19) | Rail, Barge, None | None | None | None | Fluid Bed Combustion System |
| (20) through (38) Repeat above options with underground mined Eastern Coal | | | | | |
| (39) Surface Mine<br>(Western Coal) | None | None | None | None | Conv. Boiler & Long Dist. Transmission |
| (40) Surface Mine<br>(Western Coal) | Rail | None | None | None | Conventional Boiler |
| (41) Surface Mine<br>(Western Coal) | None | Gasify (High Btu) | None | Pipeline Gas | Space Heating |
| (42) Surface Mine<br>(Western Coal) | Rail | Gasify (Low Btu) | None | None | Conventional Boiler |
| | | **Fuel/Energy Systems - Gas** | | | |
| (1) Gas Well<br>(2) (Continental)<br>(3) | None | Desulfurization | None | Gas Pipeline | Conv. Boiler,<br>Space Heating,<br>Combined Cycle |
| (4) Gas Well<br>(5) (Offshore U.S)<br>(6) | None | Desulfurization | None | Gas Pipeline | Conv. Boiler,<br>Space Heating,<br>Combined Cycle |
| (7) Gas Well<br>(8) (Arctic)<br>(9) | None | Desulfurization | None | Arctic Gas Pipeline | Conv. Boiler,<br>Space Heating,<br>Combined Cycle |
| (10) Gas Well<br>(11) (Overseas)<br>(12) | None | Desulfurization | None | Cryogenic Tanker & Stg. | Conv. Boiler<br>Space Heating<br>Combined Cycle |

Table 3. (Continued)

| Extraction Process | Transport | Processing of Raw Fuel | Secondary Processing | Transport & Storage | Utilization |
|---|---|---|---|---|---|
| Fuel/Energy Systems - Oil | | | | | |
| (1) Oil/Gas Well<br>(2) (Continental) | Oil Pipeline | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Conventional Boiler |
| (3) Oil/Gas Well<br>(4) (Offshore) | Oil Pipeline | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Conventional Boiler |
| (5) Oil/Gas Well<br>(6) (Overseas) | Tanker | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Conventional Boiler |
| (7) Oil/Gas Well<br>(Continental)<br>(8) (Offshore) | Oil Pipeline | U.S. Refinery | No. 2 Oil | Pipeline | Space Heating |
| (9) Oil/Gas Well<br>(Foreign) | Tanker | U.S. Refinery | No. 2 Oil | Pipeline | Space Heating |
| (10) Oil/Gas Well<br>(Continental)<br>(11) (Offshore) | Oil Pipeline | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Fluid Bed Combustion |
| (12) Oil/Gas Well<br>(Continental)<br>(13) (Offshore) | Oil Pipeline | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Conv. Boiler with Flue Gas Cleaning |
| (14) Oil/Gas Well<br>(15) (Foreign) | Tanker | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Fluid Bed Combustion |
| (16) Oil/Gas Well<br>(17) (Foreign) | Tanker | Topping Refinery | No. 2 Oil | Barge, Tanker | Space Heating |
| (18) Oil/Gas Well<br>(19) (Foreign) | Tanker | Topping Refinery | No. 2 Oil | Barge, Tanker | Combined Cycle |
| (20) Oil/Gas Well<br>(Foreign) | Tanker | Topping Refinery | Light Oil Gasification | Gas Pipeline | Space Heating |
| (21) Oil/Gas Well<br>(22) (Foreign) | Tanker | Topping Refinery | Naphtha Production | Barge, Tanker | Combined Cycle |
| (23) Oil/Gas Well<br>(24) (Foreign) | Tanker | Topping Refinery | Resid. Desulf. | Barge, Tanker | Conventional Boiler |
| (25) Oil/Gas Well<br>(26) (Foreign) | Tanker | Topping Refinery | Blending | Barge, Tanker | Conventional Boiler |
| (27) Oil/Gas Well<br>(28) (Foreign) | Tanker | Topping Refinery | None - Hi S Resid. | Barge, Tanker | Conv. & Flue Gas Cleaning |
| (29) Oil/Gas Well<br>(30) (Foreign) | Tanker | Topping Refinery | None - Hi S Resid. | Barge, Tanker | Fluid Bed Combustor |
| (31) Oil/Gas Well<br>(Foreign) | Tanker | Topping Refinery | Heavy Oil Gasification | Gas Pipeline | Space Heating |

## Table 3. (Continued)

| Extraction Process | Transport | Processing of Raw Fuel | Secondary Processing | Transport & Storage | Utilization |
|---|---|---|---|---|---|
| | | Fuel/Energy Systems - Oil | | | |
| (1) Oil/Gas Well<br>(2) (Continental) | Oil Pipeline | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Conventional Boiler |
| (3) Oil/Gas Well<br>(4) (Offshore) | Oil Pipeline | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Conventional Boiler |
| (5) Oil/Gas Well<br>(6) (Overseas) | Tanker | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Conventional Boiler |
| (7) Oil/Gas Well<br>(Continental)<br>(8) (Offshore) | Oil Pipeline | U.S. Refinery | No. 2 Oil | Pipeline | Space Heating |
| (9) Oil/Gas Well<br>(Foreign) | Tanker | U.S. Refinery | No. 2 Oil | Pipeline | Space Heating |
| (10) Oil/Gas Well<br>(Continental)<br>(11) (Offshore) | Oil Pipeline | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Fluid Bed Combustion |
| (12) Oil/Gas Well<br>(Continental)<br>(13) (Offshore) | Oil Pipeline | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Conv. Boiler with Flue Gas Cleaning |
| (14) Oil/Gas Well<br>(15) (Foreign) | Tanker | U.S. Refinery | Resid. Desulf. | Barge, Tanker | Fluid Bed Combustion |
| (16) Oil/Gas Well<br>(17) (Foreign) | Tanker | Topping Refinery | No. 2 Oil | Barge, Tanker | Space Heating |
| (18) Oil/Gas Well<br>(19) (Foreign) | Tanker | Topping Refinery | No. 2 Oil | Barge, Tanker | Combined Cycle |
| (20) Oil/Gas Well<br>(Foreign) | Tanker | Topping Refinery | Light Oil Gasification | Gas Pipeline | Space Heating |
| (21) Oil/Gas Well<br>(22) (Foreign) | Tanker | Topping Refinery | Naphtha Production | Barge, Tanker | Combined Cycle |
| (23) Oil/Gas Well<br>(24) (Foreign) | Tanker | Topping Refinery | Resid. Desulf. | Barge, Tanker | Conventional Boiler |
| (25) Oil/Gas Well<br>(26) (Foreign) | Tanker | Topping Refinery | Blending | Barge, Tanker | Conventional Boiler |
| (27) Oil/Gas Well<br>(28) (Foreign) | Tanker | Topping Refinery | None - Hi S Resid. | Barge, Tanker | Conv. & Flue Gas Cleaning |
| (29) Oil/Gas Well<br>(30) (Foreign) | Tanker | Topping Refinery | None - Hi S Resid. | Barge, Tanker | Fluid Bed Combustor |
| (31) Oil/Gas Well<br>(Foreign) | Tanker | Topping Refinery | Heavy Oil Gasification | Gas Pipeline | Space Heating |

Table 4. Selected Modules Analyzed

Strip Mining of Eastern Coal
Strip Mining of Western Coal
Deep Mining of Coal
Physical Cleaning of Coal
Chemical Cleaning of Coal
Liquefaction of Coal (Solvent Refining)
Rail Transport of Coal
Conventional Boiler, Eastern Coal
Conventional Boiler, Western Coal
Conventional Boiler, Physically Cleaned Eastern Coal
Conventional Boiler, Chemically Cleaned Eastern Coal
Conventional Boiler, Liquefied Coal
Conventional Boiler (Limestone Scrubber), Eastern Coal
Conventional Boiler (MgO Scrubber), Eastern Coal
Conventional Boiler (Limestone Scrubber), Western Coal
Conventional Boiler (Limestone Scrubber), Physically Clean Eastern Coal
Gasification (Low Btu) Eastern Coal - Lurgi
Gasification (High Btu) Eastern Coal - Hygas
Gasification (High Btu) Lignite - $CO_2$ Acceptor
Gasification (Low Btu) Eastern Coal - Molten Iron Combustion
Gasification of Crude Oil
High Pressure Fluidized Bed
Chemically Active Fluidized Bed
Gas Well
Gas Desulfurization
Gas Pipeline
Conventional Boiler, Natural Gas
Underground Gas Storage
LNG Tanker
LNG Port Facilities
LNG Storage
LNG Distribution
LNG Gasification
Oil Shale Extraction and Processing
Oil/Gas Well - On Shore
Oil/Gas Well - Off Shore
Oil Tanker Transport
Oil Pipeline
Oil Barge
Refinery - Domestic Crude
Refinery - Imported Crude
Topping Refinery
Conventional Boiler, Domestic Residual
Conventional Boiler, Topping Residual
Municipal Refuse Processing (St. Louis Method)
Municipal Refuse Burning, Conventional Boiler, Limestone Scrubber
Space Heating - Electrical, gas, oil, coal and synthetic gas from coal
Nuclear Fission

### *Derivation of Emission Values*

The pollutant emissions resulting from the operation involved within each module were derived from various sources. These values were then reduced to a common basis, namely, emission per million Btu. This reference energy was taken as the heat value of the primary fuel produced by the module, except in the case of electric power generation, for which the input energy to the power plant was taken as reference. Thus, in general, the emissions are given in pounds per million Btu. The land-use burden includes the land area involved and a time factor. For the processing and utilization modules, the proper unit arises from the fact that a certain land area is associated with a plant with a stated throughput, say tons of coal per hour or the equivalent heating value (Btu per hour). When the area in acres is divided by this energy rate, the resulting units are acre-hour/$10^6$ Btu. For the extraction modules, the area is associated with a total energy (e.g., tons of coal per acre). The resulting burden in acres/$10^6$ Btu is converted to consistent units by multiplying the burden by the length of time assumed for the operation.

## DATA ANALYSIS

### *Environmental Burden of Modules*

The unit-basis pollutant emissions, land-use burdens, and efficiencies for 50 modules were compiled and analyzed. From this extensive data base, a summary was prepared and is given in table 5. The values are stated in pounds per million Btu for air emissions of $NO_x$, $SO_2$, and particulates, for water emissions of suspended solids and organic materials, and for solid wastes produced. The land-use burden is expressed in acre-hour per million Btu. Data are given in table 5 for selected gas, oil, and coal modules which include extraction, transportation, conversion or treatment, and utilization.

The data presented in table 5 illustrate the differences in burdens that exist among the various modules. These burdens arise from the different steps in the fuel-energy cycle, and the various components of the environment can be affected. It is difficult to evaluate alternative energy systems directly on the basis of emissions data as presented in table 5. Therefore, it was believed that it was desirable to aggregate these complex emissions at various levels and to compile them into a single number which will

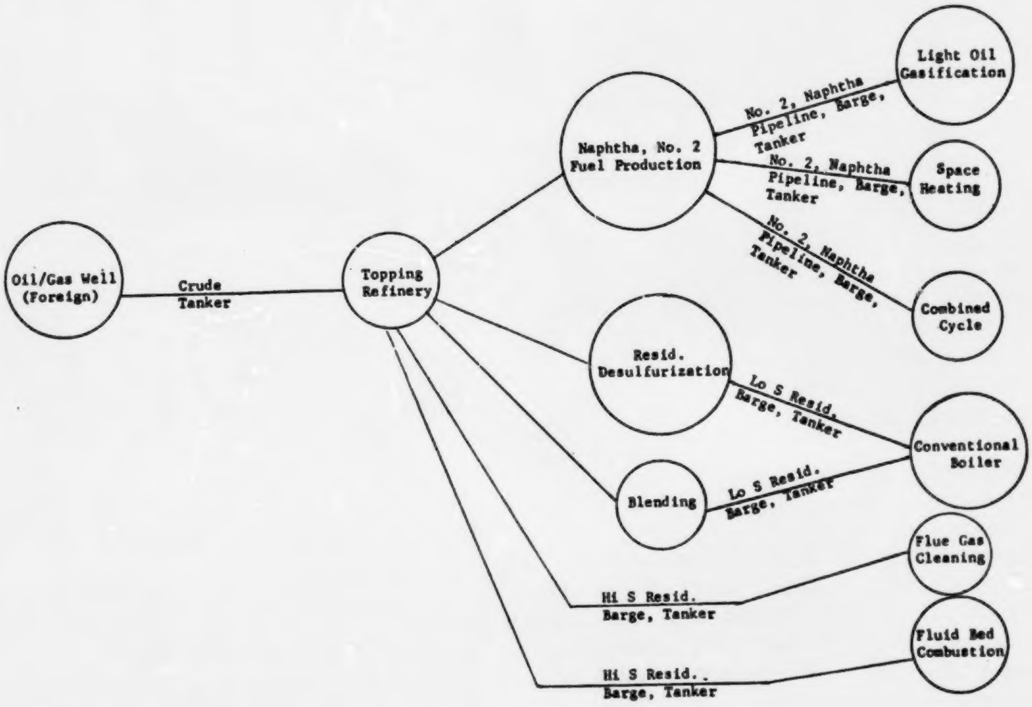
Oil/Gas Well (Foreign)
Crude Tanker
Topping Refinery
Naphtha, No. 2 Fuel Production
No. 2, Naphtha Pipeline, Barge, Tanker
Light Oil Gasification
No. 2, Naphtha Pipeline, Barge, Tanker
Space Heating
No. 2, Naphtha Pipeline, Barge, Tanker
Combined Cycle
Resid. Desulfurization
Lo S Resid. Barge, Tanker
Conventional Boiler
Blending
Lo S Resid. Barge, Tanker
Flue Gas Cleaning
Hi S Resid. Barge, Tanker
Fluid Bed Combustion
Hi S Resid. Barge, Tanker

Figure 4. Schematic Representation of Modular Relationships - Oil, Modified

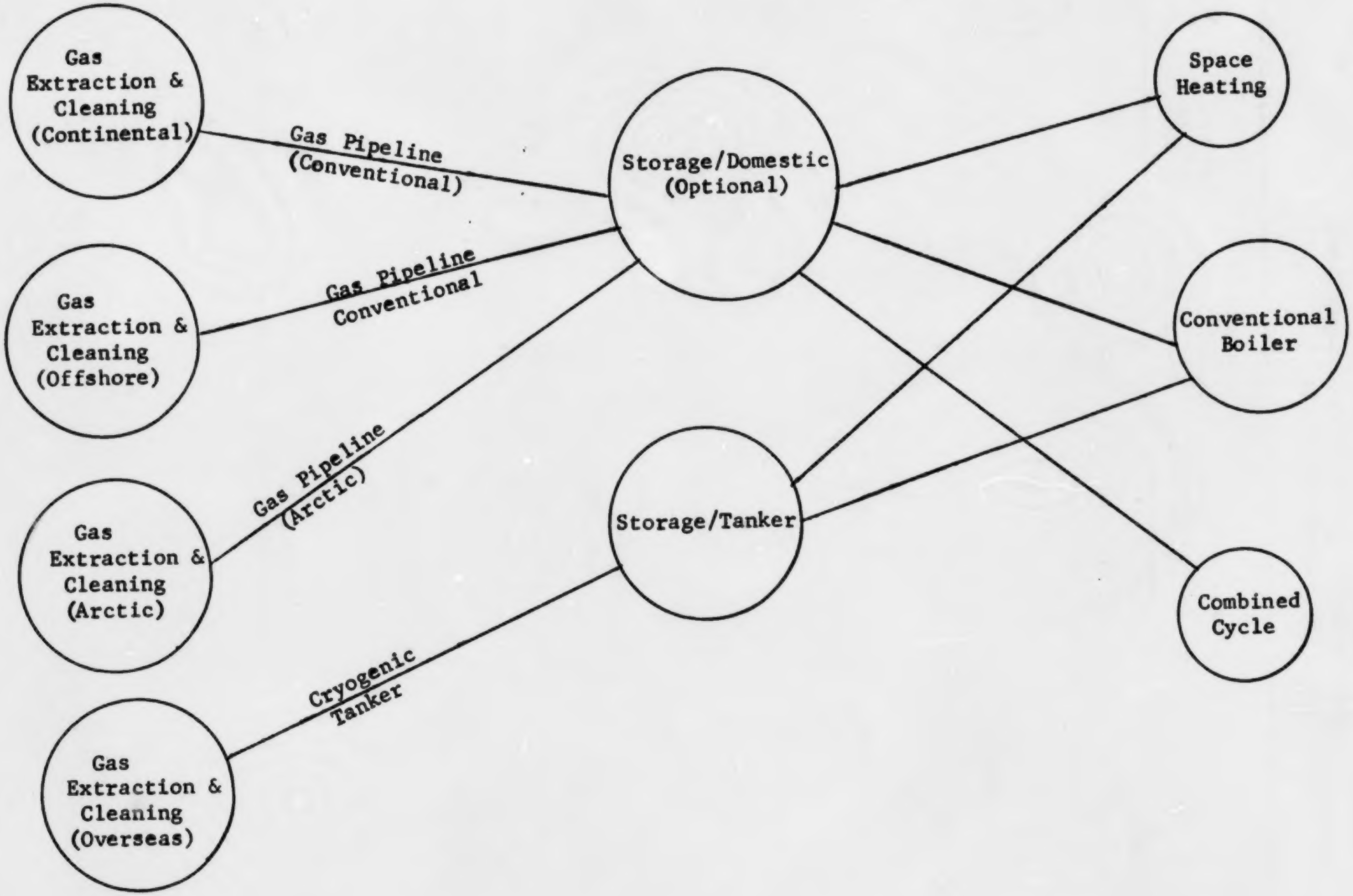
Gas Extraction & Cleaning (Continental)
Gas Extraction & Cleaning (Offshore)
Gas Extraction & Cleaning (Arctic)
Gas Extraction & Cleaning (Overseas)
Gas Pipeline (Conventional)
Gas Pipeline Conventional
Gas Pipeline (Arctic)
Cryogenic Tanker
Storage/Domestic (Optional)
Storage/Tanker
Space Heating
Conventional Boiler
Combined Cycle

Figure 5. Schematic Representation of Modular Relationships -- Gas

## Table 5. Summary of Principal Environmental Burdens for Some Selected Modules

|  | Air Emissions, lb/$10^6$ Btu |  |  | Water Emissions, lb/$10^6$ Btu |  | Solid Waste, lb/$10^6$ Btu |  | Land Use, acre-hour per $10^6$ Btu |
|---|---|---|---|---|---|---|---|---|
| Selected Modules | $NO_x$ | $SO_2$ | Particulates | Suspended Solid | Organic | Ash | Sludge |  |
| Extraction Modules |  |  |  |  |  |  |  |  |
| Strip mining-Eastern (E.) coal | 0.0002 | Neg | 0.14 | 0.55 | Neg | 0 | 0.24 | 0.3 |
| Strip mining-Western (W.) coal | 0.00008 | Neg | 0.07 | 0.28 | Neg | 0 | 0 | 0.16 |
| Onshore oil well | $8 \times 10^{-6}$ | $6 \times 10^{-5}$ | $3 \times 10^{-6}$ | 0 | 0.008 | 0 | 0 | 0.06 |
| Natural gas well | 0.23 | Neg | Neg | 0 |  | 0 | 0 | 0.06 |
| Transportation Modules |  |  |  |  |  |  |  |  |
| Rail-coal | 0.02 | 0.0014 | 0.0014 | Neg | Neg | 0.083 | Neg | 0.29 |
| Pipeline-oil | 0.009 | 0.016 | 0.002 | 0 | 0 | 0 | 0 | 0.3 |
| Pipeline-gas | 0.304 | 0 | 0 | 0 | 0 | 0 | 0 | 1.0 |
| Conversion or Treatment Modules |  |  |  |  |  |  |  |  |
| Physical coal cleaning | 0.006 | 0.004 | 0.01 | Neg | Neg | 0 | 0.3 | 0.003 |
| Chemical coal cleaning | 0.04 | 0.1 | 0.005 | Neg | Neg | 0 | 0 | 0.08 |
| Solvent refining of coal | 0.21 | 0.003 | 0.27 | 0 | Neg | 16. | 0 | 0.08 |
| Refining of oil-domestic crude | 0.025 | 0.135 | 0.002 | 0.004 | 0.002 | 0 | 0.026 | 0.009 |
| Natural gas desulfurization | 0 | 0.0183 | 0 | Neg | Neg | Neg | Neg | 0.005 |
| Utilization Modules |  |  |  |  |  |  |  |  |
| Conventional boiler(CB)-W. coal | 0.98 | 1.65 | 0.07 | 0.025 | 0.011 | 9. | 0 | 0.1 |
| C.B. with limestone scrubber (LSS)-W. coal | 0.78 | 0.16 | 0.07 | 0.025 | 0.011 | 1.8 | 13.4 | 0.1 |
| Fluidized-bed combustion plus combined cycle-E. coal | 0.14 | 0.7 | 0.02 | 0 | 0 | 17.3 | 0 | 0.12 |
| Gasification (molten iron combustion) plus C.B.-E. coal | 0.39 | 0.017 | 0.034 | 0 | 0 | 10. | 0 | 0.12 |
| C.B. with MgO scrubber-E. coal | 0.60 | 0.50 | 0.1 | 0.025 | 0.011 | 2.4 | 10.4 | 0.1 |
| C.B.-physically cleaned E. coal | 0.68 | 2.02 | 0.044 | 0.025 | 0.011 | 5.4 | 0 | 0.1 |
| C.B. with LSS-physically cleaned E. coal | 0.55 | 0.2 | 0.044 | 0.025 | 0.011 | 1.1 | 11.9 | 0.1 |
| C.B.-chemically cleaned E. coal | 0.75 | 1.93 | 0.1 | 0.025 | 0.011 | 11.9 | 0 | 0.1 |
| C.B.-solvent refined E. coal | 0.56 | 0.71 | 0.0003 | 0.025 | 0.011 | 0.031 | 0 | 0.09 |
| Chemically active fluidized-bed plus combined residual oil | 0.16 | 0.45 | 0.01 | 0 | 0 | 3.0 | 0 | 0.06 |
| C.B.-residual oil from domestic crude | 0.70 | 1.83 | 0.05 | 0 | 0 | 0 | 0 | 0.04 |
| C.B.-natural gas | 0.39 | 0.0006 | 0.015 | 0.016 | 0 | 0 | 0 | 0.02 |

reflect all of the emissions, thus aiding in comparing the environmental aspects of various energy systems.

A method for accomplishing the aggregation of emissions, based upon methodology developed for the Bureau of Reclamation, U.S. Department of the Interior, was employed (ref. 1). The details of the ranking methodology are presented in the Appendix. During this brief study, no attempt was made to detail the analysis on a regional basis. The results thus represent a national overview which can be useful in the evaluation of national priorities. Ultimate implementation of energy policy should include consideration of regional factors.

The first level of aggregation of the emission data is the summation of the weighted parameters for each environmental component within the module. The resulting totals for the air, water, solid, and land-use burdens are compiled for each module in table 6. Since the actual emissions have been weighted in the calculation of the totals given, units are not assigned to the totals. Examination of table 6 shows the range of values which occurs for a given type of burden. Some general points regarding control technology associated with the modules given in table 6 should be noted:

- Mine modules assume land restoration (80 percent coverage, no bare areas greater than ¼ acre, and 600 living stems per acre), and treatment of acid mine drainage;
- Physical coal cleaning assumes restoration of land used for refuse piles (all pyritic material covered with nonreactive soil) and treatment of acid-bearing runoff;
- Boiler modules assume 99 percent efficiency for particulate removal;
- Stack gas cleaning modules assume 90 percent reduction in $SO_2$ and 20 percent reduction in $NO_x$;
- Cooling towers are assumed for all modules discharging heat in water effluents. The heat discharged to air is not included in the burden totals.

The compilation of table 6 also shows the cross-media effects of alternate pollution control approaches. For example, Modules 8 through 16 are all conventional boilers burning several different kinds of coal, either with or without stack-gas cleaning. Comparison of Module 8 with Module 13 shows an 80 percent reduction in air burden when a limestone scrubbing system is employed, with an attendant increase in solid burden due to the gypsum sludge produced. Module 14 shows the same reduction in air burden, but it does not have the increase in solid waste, since the MgO scrubber is a regenerative system. Module 22, high-pressure fluidized-bed combustion, also is a coal utilization module which exhibits yet an even smaller air burden with an intermediate solid impact.

Thus, table 6 can be used to compare alternative processes on a burden-by-burden basis.

### *Environmental Burden of Systems*

The second level of aggregation of the emission data is the summation of the weighted totals for each environmental component over all of the modules in each chosen system. Again, weighting factors reflecting the relative importance of each module are employed in the summation. The result is a separate total for air, water, solid waste, and land-use burdens which reflect the individual contributions of all of the burdens.

The final aggregation of the emission data is the summation of the environmental component totals to give a single-system environmental index. The results obtained by this approach are described in the following paragraphs. It should be noted that this is simply one method of aggregating the complex emission data, and the results are not a unique representation of that data. The method includes the flexibility to allow any system evaluator to select weighting factors at each level of aggregation to reflect his understanding of the relative importance of each factor involved. The computer program readily allows recalculation of the system environmental indices on the basis of refined weighting factors.

During this short study, a group of 26 systems for producing electric power and a group of 5 systems for space heating were selected for analysis. The electric power group included 15 coal, 2 mixed fuel (coal plus municipal refuse), 6 oil, 1 nuclear fission, and 2 natural gas systems. These include the important options which must be considered in arriving at an energy policy. The group of space heating systems include electricity, natural gas, oil, and coal.

### *Electric Power Systems*

All of the systems which have been analyzed are listed in table 7. For each electric power system and appropriate modules, burdens given in table 6 were employed to derive an overall system environmental index. Since the module burdens are stated on a unit basis, the efficiencies must be factored in. The weighted sums in each module were first adjusted by dividing each of the four values by the product of the efficiencies of each module following it in the system sequence. The impacts for electric-power-generation

Table 6. Summary of Module Impacts

| Module Number | Module | Impacts, Weighted Sums (EWpQp) for Each Environmental Component Air | Water | Solid | Land Use |
|---|---|---|---|---|---|
| 1 | Eastern Coal, Strip Mine | 0.186 | 0.0038 | 0.240 | 0.34 |
| 2 | Western Coal, Strip Mine | 0.0933 | 0.0013 | 0.040 | 0.17 |
| 3 | Eastern Coal, Deep Mine | 0.0005 | 0.045 | 9.60 | 0.20 |
| 4 | Physical Cleaning of Coal | 0.0218 | 0 | 0.06 | 0.003 |
| 5 | Chemical Cleaning of Coal | 0.212 | 0 | 0 | 0.002 |
| 6 | Liquefaction of Coal (Solvent Refining) | 0.222 | 0 | 16.0 | 0.027 |
| 7 | Rail Transport of Coal | 0.0247 | 0 | 0.083 | 0.29 |
| 8 | Conv. Boiler, Eastern Coal | 7.040 | 0.0039 | 12.0 | 0.10 |
| 9 | Conv. Boiler, Western Coal | 3.150 | 0.0039 | 9.0 | 0.13 |
| 10 | Conv. Boiler, Physically Cleaned Eastern Coal | 3.338 | 0.0039 | 5.41 | 0.10 |
| 11 | Conv. Boiler, Chemically Cleaned Eastern Coal | 4.215 | 0.0039 | 11.41 | 0.10 |
| 12 | Conv. Boiler, Liquefied Coal | 1.585 | 0.0039 | 0.074 | 0.09 |
| 13 | Conv. Boiler, Limestone Scrubber, Eastern Coal | 1.396 | 0.0039 | 29.8 | 0.10 |
| 14 | Conv. Boiler, MgO Scrubber, Eastern Coal | 1.394 | 0.0039 | 12.16 | 0.10 |
| 15 | Conv. Boiler, Limestone Scrubber, Western Coal | 1.453 | 0.0039 | 21.05 | 0.10 |
| 16 | Conv. Boiler, Limestone Scrubber, Physically Cleaned Eastern Coal | 0.922 | 0.0039 | 13.80 | 0.10 |

Table 6. (Continued)

| Module Number | Module | Impacts, Weighted Sums (EWpQp) for Each Environmental Component: Air | Water | Solid | Land Use |
|---|---|---|---|---|---|
| 17 | Gasification, Eastern Coal, Lurgi (Low Btu) plus Conv. Boiler | 1.624 | 0.0046 | 9.75 | 0.12 |
| 18 | Gasification, Eastern Coal, Hygas (High Btu) | 1.79 | 0.0023 | 31.9 | 0.02 |
| 19 | Gasification, Lignite, $CO_2$ Acceptor (High Btu) | 0.027 | 0.0023 | 39.2 | 0.02 |
| 20 | Gasification, Eastern Coal, Molten Iron Combustion (Low Btu) plus C. B. | 0.449 | 0 | 10.0 | 0.12 |
| 21 | Gasification of Crude Oil | 0.17 | 0.0002 | 0.18 | 0.04 |
| 22 | High Pressure Fluidized Bed plus C. C. | 1.042 | 0 | 17.40 | 0.10 |
| 23 | Chemically Active Fluidized Bed plus C. C. | 0.761 | 0 | 3.0 | 0.12 |
| 24 | Natural Gas Well | 0.845 | 0.0031 | 0 | 0.05 |
| 25 | Natural Gas Desulfurization | 0.0229 | 0 | 0 | 0.005 |
| 26 | Gas Pipeline | 0.304 | 0 | 0 | 1.00 |
| 27 | Underground Gas Storage | 0.60 | 0 | 0 | Neg. |
| 28 | Conv. Boiler, Natural Gas | 0.446 | 0.0002 | 0 | 0.02 |
| 29 | LNG Tanker | 0 | 0 | 0 | 0 |
| 30 | LNG Port Facilities | 0 | 0 | 0 | Neg |
| 31 | LNG Storage | 0 | 0 | 0 | 0.00005 |
| 32 | LNG Gasification | 0.0026 | 0 | 0 | 0.00004 |
| 33 | Oil Shale Extraction and Processing | 0.484 | 0.001 | >400 | N.D. |

Table 6. (Continued)

| Module Number | Module | Impacts, Weighted Sums (EWpQp) for Each Environmental Component | | | |
|---|---|---|---|---|---|
| | | Air | Water | Solid | Land Use |
| 34 | Oil/Gas Well - On Shore | 0.00008 | 0.0223 | 0 | 0.05 |
| 35 | Oil/Gas Well - Off Shore | 0.00016 | 0.0023 | 0 | 0.05 |
| 36 | Oil Tanker Transport | 0.1416 | 0.0495 | 0 | 0 |
| 37 | Oil Pipeline | 0.157 | 0.00495 | 0 | 0.31 |
| 38 | Oil Barge | 0.0143 | 0.00512 | 0 | 0 |
| 39 | Refinery, Domestic Crude | 0.214 | 0.0068 | 0.026 | 0.009 |
| 40 | Refinery, Imported (Arabic) Crude | 0.225 | 0.0068 | 0.026 | 0.009 |
| 41 | Topping Refinery | 0.155 | 0.00425 | 0.026 | 0.009 |
| 42 | Conv. Boiler, Domestic Resid. | 3.161 | 0 | 0 | 0.04 |
| 43 | Conv. Boiler, Topping Resid. | 1.811 | 0 | 0 | 0.04 |
| 44 | Municipal Refuse Processing (St. Louis Method) | 0 | 0 | 0 | 0.034 |
| 45 | Municipal Refuse Burning, Conv. Boiler, Limestone Scrubbing | 0.717 | 0 | -132.0 | 0.1 |
| 46 | Space Heating, Electricity | 0 | 0 | 0 | 0 |
| 47 | Space Heating, Gas | 0.572 | 0 | 0 | 0 |
| 48 | Space Heating, Oil | 1.13 | 0 | 0 | 0 |
| 49 | Space Heating, Coal | 10.5 | 0 | 0 | 0 |
| 50 | Nuclear Fission | 0.0032 | 0.022 | 4.0 | 0.3 |

modules are stated on a unit input basis; therefore, the efficiency factor is applied to those impacts as well, in order to put all systems on a common output basis. The adjusted burdens of all modules in a given system were then added to give total air, water, solid, and land-use burdens. These totals are given for each system in table 7. Again, the burden data in table 7 are useful in comparing the tradeoffs which are involved in the consideration of various system options.

The total air, water, solid, and land-use values were normalized and then summed to give the overall system environmental index. The systems are arranged in ranked order in table 7 according to the value of the derived system index.

Three additional elements are included in table 7; the overall system efficiency (the product of the efficiencies of each module in the system), the estimated overall cost to produce electricity, and the approximate year of availability of each system.

It should be emphasized that the system index as given in table 7 was derived with equal weight being given to the burdens from each module in the system and also to the air, water, solid, and land-use burdens. With these assumptions, three of the four systems which include limestone scrubbing are ranked below the corresponding system without scrubbing. This occurs because the $SO_2$ and sludge produced (16 lb sludge per lb of $SO_2$ removed) are given equal environmental importance. In view of the fact that air emissions are, in general, more likely to produce adverse health effects than are solid wastes, it would be reasonable to assign a lesser weight to solid waste in the system analysis. Based on this premise, a second calculation was carried out in which the solid burdens were assigned a weighting factor of 0.3 while keeping the remaining media weights equal to unity. The results of this calculation are given in table 8 in which the systems are ordered according to their revised ranking. For comparison, the rank of each system obtained previously with equal media weights also is given. The revised order shows the expected changes. Systems with relatively large solid burdens moved up in rank. For example, the systems which include limestone scrubbing are ranked above the corresponding systems without scrubbing.

## DISCUSSION OF THE RESULTS

### *Systems Relationships*

Inspection of tables 7 and 8 reveals a number of interesting features. A few of these may be noted as illustrative of the comparisons which may be drawn:

- Natural gas and LNG systems predictably rank high environmentally;
- Among the currently available systems, nuclear fission, western coal, and imported oil-topping refinery systems are highly ranked;
- MgO scrubbing systems and, given the assumption of lesser weight for solid waste, limestone scrubbing systems are favorably ranked;
- Various coal cleaning and processing options show significant environmental gains over the use of untreated coal;
- The developmental technologies: chemically active fluidized-bed combustion of oil, molten iron gasification of coal, and high-pressure fluidized-bed combustion of coal, are highly promising from the environmental standpoint;
- Systems, including strip-mining of coal, are environmentally acceptable if land restoration and treatment of acid-mine drainage is practiced;
- The oil systems are intermediate in rank due to higher than average air and water emissions.

One of the most striking results is the high ranking of the coal-municipal refuse, mixed-fuel systems. This option is highly promising from a number of viewpoints. The modules (processing and burning of municipal refuse) are based on the St. Louis approach (arbitrarily selected for illustration), and the system burdens were derived by combining the appropriate fraction of the burdens from the modules involved. This option consumes the combustible portion of the municipal refuse which obviates the need for an alternate means of disposal and, in turn, results in a negative value (a credit) for the solid waste burden of the module. In addition, the prepared refuse replaces a portion, 10 to 20 percent in the cases included, of the coal required by the power plant, thus reducing the coal system burden by that fraction.

### *Systems Availability*

An approximate picture of the estimated availability time frame may be drawn by listing the systems as follows:

(1) Currently available
    LNG
    Nuclear fission
    Natural gas
    Oil-conventional boiler
    Coal-physical cleaning
(2) Available by 1975
    Mixed fuel-coal-municipal refuse
    Topping of imported crude oil
    Limestone scrubbing of flue gas

## Table 7. Summary of Ranked Systems, with Media Impacts, Efficiencies, and Costs (Weighting Factors for Air, Water, Solid, and Land Uses =1)

| Rank | System Environ. Index | System | | | | | Total Air Impact | Total Water Impact | Total Solid Impact | Total Land Impact | Overall System Eff. | Overall* Cost, mills/kwh | Est. Year of Application |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Extraction | Transportation | Processing | Transportation | Utilization | | | | | | | |
| 1 | 1.5 | None | LNG Tanker/Storage/Distribution/Gasification | | | Conv. boiler | 1.21 | 0.0004 | 0 | 0.054 | .323 | | Current |
| 2 | 5.2 | None | Imported resid | Fl. bed combust. | | Combined cycle | 2.00 | 0 | 7.89 | 0.315 | .390 | 11.3 | 1985 |
| 3 | 11.2 | 80% E. coal-20% Prepared Mun. Refuse | | | | Conv. boiler lime scrub | 4.08 | 0.018 | -6.58 | 1.75 | .348 | 11.5 | 1975 |
| 4 | 12.4 | | Nuclear Fission - Total System** | | | | 0.0032 | 0.022 | 4. | 0.3 | .300 | 11.0 | Current |
| 5 | 16.7 | Nat. gas well | | Desulf. nat. gas | Nat. gas pipeline | Conv. boiler | 4.47 | 0.0092 | 0 | 2.91 | .341 | 9.1 | Current |
| 6 | 19.7 | East coal, S.M. | Rail | Gasify, low Btu molt. iron combust. | None | Conv. boiler | 2.00 | 0.0115 | 31.28 | 2.27 | .329 | | 1985 |
| 7 | 22.6 | West coal, S.M. | Rail | None | None | Conv. boiler | 8.83 | 0.0140 | 24.66 | 1.59 | .369 | 11.5 | Current |
| 8 | 23.3 | East coal, S.M. | Rail | Fl. bed combust. | None | Combined cycle | 3.00 | 0.0100 | 46.64 | 1.92 | .378 | 10.8 | 1965 |
| 9 | 23.7 | 90% E. coal-10% Prepared Mun. Refuse | | | | Conv. boiler lime scrub | 4.34 | 0.02 | 39.7 | 1.92 | .348 | 11.5 | 1975 |
| 10. | 23.9 | East coal, S.M. | Rail | None | None | Conv. boiler MgO scrub | 4.59 | 0.0220 | 35.67 | 2.09 | .349 | 11.5 | 1980 |
| 11 | 24.6 | None | Oil tanker | Topping refinery | Oil barge | Conv. boiler topp. resid. | 5.75 | 0.163 | 0.0703 | 0.132 | .324 | | 1975 |
| 12 | 24.6 | East coal | Rail | Physical coal clean | None | Conv. boiler | 9.73 | 0.0221 | 15.76 | 2.21 | .324 | 10.0 | Current |
| 13 | 25.0 | East coal | Rail | Physical coal clean | None | Conv. boiler lime scrub | 3.38 | 0.0234 | 40.65 | 2.34 | .307 | 12.4 | 1975 |
| 14 | 27.1 | West coal, S.M. | Rail | None | None | Conv. boiler lime scrub | 4.49 | 0.0148 | 60.49 | 1.58 | .349 | 13.3 | 1975 |
| 15 | 27.5 | Oil well, onshore | Oil pipeline | Refinery domestic | Oil barge | Conv. boiler dom. resid. | 9.64 | 0.1165 | 0.0703 | 1.24 | .322 | 11.2 | Current |
| 16 | 29.3 | East coal, S.M. | Rail | Coal liquefaction | None | Conv. boiler | 5.64 | 0.0242 | 44.61 | 2.58 | .276 | 13.2 | 1985 |
| 17 | 30.7 | East coal, S.M. | Rail | Chemical coal clean | None | Conv. boiler | 12.57 | 0.0213 | 31.76 | 2.07 | .350 | 10.5 | 1980 |
| 18 | 30.8 | Oil well, onshore | Oil pipeline | Crude oil gasification | Nat. gas pipeline | Conv. boiler | 3.09 | 0.1016 | 0.507 | 4.20 | .270 | | 1980 |
| 19 | 33.2 | East coal, S.M. | Rail | Gasif. low Btu Lurgi. | None | Conv. boiler | 7.49 | 0.0342 | 41.11 | 3.06 | .244 | | 1985 |
| 20 | 34.4 | Oil well, off-shore | Oil pipeline | Refinery domestic | Oil tanker | Conv. boiler dom. resid. | 9.99 | 0.174 | 0.071 | 1.25 | .321 | 11.2 | Current |
| 21 | 36.3 | East coal, S.M. | Rail | None | None | Conv. boiler lime scrub | 4.59 | 0.0221 | 86.0 | 2.09 | .348 | 11.5 | 1975 |
| 22 | 37.7 | East coal, S.M. | Rail | None | None | Conv. boiler | 19.6 | 0.0207 | 33.31 | 1.97 | .369 | 9.1 | Current |
| 23 | 41.3 | Oil well, onshore | Oil pipeline | Refinery domestic | Oil tanker | Conv. boiler dom. resid. | 9.99 | 0.237 | 0.071 | 1.25 | .321 | 11.2 | Current |
| 24 | 43.6 | East coal, D.M. | Rail | Physical coal clean | None | Conv. boiler | 9.16 | 0.148 | 44.49 | 1.78 | .324 | 10.0 | Current |
| 25 | 45.1 | East coal, D.M. | Rail | Physical coal clean | None | Conv. boiler lime scrub | 2.78 | 0.157 | 71.04 | 1.89 | .307 | 12.4 | 1975 |
| 26 | 53.3 | East coal, D.M. | Rail | None | None | Conv. boiler | 19.09 | 0.1216 | 58.60 | 1.59 | .369 | 9.1 | Current |

* Approximate-system costs may vary by ± 2.5 mills/kWhr for specific cases.

** Includes only environmental impacts resulting from normal controlled operation at this time. Analysis of nuclear accident probability and effect is not complete.

## Table 8. Alternate Ranking of Systems

Weighting Factor for Solid Waste = 0.3
Weighting Factors for Air, Water, and Land Use = 1

| Rank for $W_s = 0.3$ | System Environmental Index | Rank for All $W_n = 1$ | System | | | | |
|---|---|---|---|---|---|---|---|
| | | | Extraction | Transportation | Processing | Transportation | Utilization |
| 1 | 1.5 | 1 | None | LNG/Tanker/Storage/Distribution/Gasification | | | Conv. boiler |
| 2 | 3.8 | 2 | None | Imported resid | Fl. bed combust. | | Combined cycle |
| 3 | 12.2 | 4 | Nuclear Fission - Total System* | | | | |
| 4 | 12.3 | 3 | 80% E. coal-20% Prepared Mun. Refuse | | | | Conv. boiler lime scrub |
| 5 | 14.4 | 6 | East coal, S.M. | Rail | Gasify, low Btu molt. iron combust. | None | Conv. boiler |
| 6 | 15.3 | 8 | East coal, S.M. | Rail | Fl. bed combust. | None | Combined cycle |
| 7 | 16.7 | 5 | Nat. gas well | | Desulf. nat. gas | Nat. gas pipeline | Conv. boiler |
| 8 | 16.7 | 14 | West coal, S.M. | Rail | None | None | Conv. boiler lime scrub |
| 9 | 16.9 | 9 | 90% E. coal-10% Prepared Mun. Refuse | | | | Conv. boiler lime scrub |
| 10 | 17.7 | 10 | East coal, S.M. | Rail | None | None | Conv. boiler MgO scrub |
| 11 | 18.0 | 13 | East coal | Rail | Physical coal clean | None | Conv. boiler lime scrub |
| 12 | 18.4 | 7 | West coal, S.M. | Rail | None | None | Conv. boiler |
| 13 | 21.5 | 21 | East coal, S.M. | Rail | None | None | Conv. boiler lime scrub |
| 14 | 21.6 | 16 | East coal, S.M. | Rail | Coal liquefaction | None | Conv. boiler |
| 15 | 21.9 | 12 | East coal | Rail | Physical coal clean | None | Conv. boiler |
| 16 | 24.5 | 11 | None | Oil tanker | Topping refinery | Oil barge | Conv. boiler topp. resid. |
| 17 | 25.3 | 17 | East coal, S.M. | Rail | Chemical coal clean | None | Conv. boiler |
| 18 | 26.2 | 19 | East coal, S.M. | Rail | Gasif. low Btu Lurgi. | None | Conv. boiler |
| 19 | 27.5 | 15 | Oil well, onshore | Oil pipeline | Refinery domestic | Oil barge | Conv. boiler dom. resid. |
| 20 | 30.7 | 18 | Oil well, onshore | Oil pipeline | Crude oil gasification | Nat. gas pipeline | Conv. boiler |
| 21 | 32.0 | 22 | East coal, S.M. | Rail | None | None | Conv. boiler |
| 22 | 32.9 | 25 | East coal, D.M. | Rail | Physical coal clean | None | Conv. boiler lime scrub |
| 23 | 34.4 | 20 | Oil well, offshore | Oil pipeline | Refinery domestic | Oil tanker | Conv. boiler dom. resid. |
| 24 | 35.9 | 24 | East coal, D.M. | Rail | Physical coal clean | None | Conv. boiler |
| 25 | 41.3 | 23 | Oil well, onshore | Oil pipeline | Refinery domestic | Oil tanker | Conv. boiler dom. resid. |
| 26 | 43.2 | 26 | East coal, D.M. | Rail | None | None | Conv. boiler |

* Includes only environmental impacts resulting from normal controlled operation at this time. Analysis of nuclear accident probability and effect is not complete.

(3) Available by 1980
    MgO scrubbing of flue gas
    Chemical coal cleaning
    Crude oil gasification
(4) Available by 1985
    Fluidized-bed combustion of coal and oil
    Gasification of coal
    Liquefaction of coal (solvent refining).

### *Cost of Pollution Control*

The costs associated with the various mechanisms for the control of emissions from different phases in the fuel-energy cycle have been developed.* A summary of the ranges of control costs in terms of medium, fuel, and module is given in table 9.

### Conclusions

(1) The major results of this study and the conclusions which have been drawn may be summarized as follows:

(a) A preliminary compilation of effluent data has been developed for those energy sources considered commercially viable in the 1975 to 1990 time period. These data represent readily available information within EPA and private industry concerning the quantity of residual pollutants produced during the extraction, conversion, transportation, and stationary use of fuels to produce electricity or direct heating under best available conditions of environmental control. The incremental cost of control and the overall cost of electricity produced also have been compiled.

(b) A preliminary methodology has been developed for organizing the effluent data collected and for combining the emission data for each module into a single environmental index for each energy system. The value of such an index is that it provides a tool for making explicit the value judgments of any system evaluator with respect to the relative environmental impact of energy systems. This methodology has been applied in the context of this study to develop a gross environmental ranking of the energy systems. This methodology has been applied in the context of this study to develop a gross environmental ranking of the energy systems considered. Ultimately, such a ranking of energy systems must be done on the basis of specific and local environmental impacts which may vary considerably the weighting factor associated with each module and pollutant.

(c) From the data compiled in this study, it is clear that natural gas systems produce electrical energy with the least associated environmental burdens. Moderate air emissions occur in the extraction and combustion phases, but other burdens are small or negligible. Electrical energy produced by residual fuel oil systems gives rise to greater environmental burdens. Significant air emissions occur in the refining and combustion phases, and water pollutants are produced in the refining phase. Eastern coal-based systems, which employ current technology, produce still greater environmental burdens, chiefly in the form of solid waste from extraction and combustion, and air emissions from combustion.

(d) The application of improved technology in the areas of fuel conversion and pollution control can be expected to achieve substantial reduction in the overall environmental burdens. The control of $SO_2$ emissions from coal and oil combustion needed to achieve ambient air quality standards can be technically achieved in the 1975 to 1990 time period. Similar conclusions may be drawn regarding coal conversion technologies such as liquefaction and gasification. However, such treatment or control technologies must transfer the inherent fuel sulfur, plus chemical reactant, to another media creating water pollutants or solid waste products. Near the end of the 1975 to 1990 time period, regenerative stack-gas-cleaning technologies, such as MgO scrubbing, and advanced combustion techniques, such as fluidized-bed combustion of coal and oil, can be made available which will achieve equivalent reduction of air emissions with only a moderate increase in the production of solid waste. This conclusion can be illustrated by comparing the approximate total annual air emissions associated with the extraction, transportation, and combustion of eastern coal to produce 1,000 MW of electricity: for coal burned in a conventional boiler, 235,000 tons; for conventional boiler plus wet limestone scrubbing, 60,000 tons; for conventional boiler plus MgO scrubbing, 60,000 tons; and for fluidized-bed combustion of coal plus combined cycle, 40,000 tons. The corresponding approximate total annual production of solid waste for the same four systems is: 500,000; 1,300,000; 530,000; and 700,000 tons; respectively. By comparison, the

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*Report referred to in "Acknowledgment" at end of this paper.

Table 9. Summary of Control Costs

| Medium | | Control Cost per $10^6$ Btu | Module |
|---|---|---|---|
| **Air** | | | |
| Coal | | \$0.10 - \$0.35 | Processing, power plant |
| Gas | | 0.01 - 0.05 | Extraction, power plant |
| Oil | | 0.05 - 0.25 | Processing, power plant |
| | Total | \$0.16 - \$0.65 | |
| **Water** | | | |
| Coal | | \$0.005 - \$0.01 | Extraction |
| Gas | | 0.005 - | Extraction |
| Oil | | 0.005 - 0.01 | Extraction, processing |
| | Total | \$0.015 - \$0.02 | |
| **Solid** | | | |
| Coal | | \$0.01 - \$0.10 | Extraction, processing, power plant |
| Gas | | 0 - 0 | |
| Oil | | 0 - 0.03 | Processing |
| | Total | \$0.01 - \$0.13 | |

total of the air emissions associated with the extraction, transportation, and combustion of natural gas is about 80,000 tons. The solid waste production for the natural gas system is negligible. Similarly, the approximate total annual air emissions from the extraction, transporation, refining, and combustion of oil could be reduced by wet limestone scrubbing of the power plant stack gas from about 120,000 tons to about 75,000 tons with an associated increased production of 322,500 tons of solid waste. Application of MgO scrubbing would achieve the same level of air emission with minimal increase of solid waste production above no scrubbing;

(e) Based on this evaluation, the air emissions from coal-based systems can be reduced by application of advanced technology to less than those of natural gas, while minimizing the attendant increase in solid waste.

(f) The data bank and computer program for the ranking procedure are extant. The computerized methodology makes it simple to test the preferred weighting factors of any energy-system evaluator.

(g) The pertinent environmental factors have been identified for those advanced energy sources considered to be developmental during the 1975 to 1990 time period. The qualitative environmental relationships have been evaluated by a panel through value judgments and subjective considerations. Advanced energy sources judged to exhibit major potential for supplying significant portions of future energy demand with reduced environmental impact are geothermal, solar, and nuclear fusion.

(2) The very broad scope of the project and the short time available for this preliminary study necessarily limited the effort to an overview. Nevertheless, the assembled data represent a unique compilation of emission inventories which can serve as a foundation and point of departure for both technical investigators and policymakers.

(3) Emission data for hazardous trace materials are grossly inaccurate or not available.

## APPENDIX
## METHODOLOGY FOR RANKING OF ENERGY SYSTEMS

### *Approach*

The evaluation of alternative systems for the production of useful energy, and the modules contained within these systems, required the comparison of a variety of environmental burdens. These burdens can arise through different steps in the process, and various components of the environment can be affected. Some system of ranking which attempts to collect these complex burdens into a single number or set of numbers is desirable as part of the evaluation scheme. An attempt has been made during this phase of the study to develop a method for the initial ranking of energy systems.

In concept, the ranking system used is based upon methodology developed for the Bureau of Reclamation, U.S. Department of the Interior (ref. 1). This concept involves a hierarchy of environmental burdens under four major environmental components, which in turn can be separated into environmental parameters. An environmental parameter is a single measurement or a series of measurements of the burden. If the environmental burden for the system or activity under consideration is to be derived properly, then it is necessary to combine the environmental burdens at each level of this hierarchy. Weighting factors are assigned as a measure of the importance of a burden at any particular level of the hierarchy and can be used to accommodate the different units used in the measurement of the various burdens.

Due to the limited time for the study, a complete hierarchy of environmental burdens was not developed. An attempt has been made to develop a quantitative system utilizing five environmental components. The components assumed to be of significance to the evaluation of energy systems are:

(1) Air,
(2) Water,
(3) Solid waste,
(4) Land use,
(5) Radiation.

Various parameters within each component were quantified as appropriate to each module. The goal of the ranking procedure used was to derive a single environmental index for the energy system under consideration. This index is represented as follows:

$$I_i = \Sigma_m W_{m_i} \Sigma_n W_n \Sigma_p W_p Q_p ,$$

where $I_i$ = Environmental index for energy system i,

$W_{m_i}$ = Weight of module m in system i,

$W_n$ = Weight of environmental component n,

$W_p$ = Weight of parameter p,

$Q_p$ = Quantity of p produced per unit of energy.

A computer code was written to facilitate the calculation of the environmental indices for the relatively large numbers of modules and for the combination of these modules into systems. The computer code also permitted the performance of sensitivity calculations to gain an appreciation of the dependence of the environmental index on uncertainties in input data. The assignment of weighting factors, the development of the computer code, and the sensitivity calculations are discussed in the following sections.

### *Derivation of Weighting Factors*

#### *Weighting Factors for Air Parameters*

The air pollution index is based on the primary emission standards and pollutant levels in terms of emissions in pounds per million Btu. The following primary standard values were employed:

| | | |
|---|---|---|
| $SO_x$ | 80 $\mu g/m^3$ | annual average value |
| $NO_x$ | 100 $\mu g/m^3$ | |
| Particulate | 75 $\mu g/m^3$ | |
| CO | 1000 $\mu g/m^3$ | annual average value extrapolation from 10,000 $mg/m^3/8$ hr |
| Hydrocarbons | 160 $\mu g/m^3$ | 1 hr—used as annual average value |
| Trace metals | | |
| Be | 0.1 $\mu g/m^3$ | |
| Hg | 1.0 $\mu g/m^3$ | |
| Pb | 2.0 $\mu g/m^3$ | |

Fine particles ($<1\mu$) and thermal emissions also should be included in the consideration of air emissions. Standard values, which could be used in the same manner as the values above, were not identified for these pollutants in this study, so these emissions were not included in the calculations of the environmental index. Their omission from the current assessment is not meant to minimize their importance, however.

The above values were referenced to the $NO_x$ value. The resulting weighting factors are:

| Pollutant | Factor ($W_p$) | Babcock's factors (normalized) |
|---|---|---|
| $NO_x$ | 1 (reference) | 514/514 = 1.00 |
| $SO_x$ | $\frac{100}{80} = 1.25$ | 514/1,430 = 0.36 |
| TSP | $\frac{100}{75} = 1.33$ | 514/375 = 1.37 |
| CO | $\frac{100}{1000} = 0.10$ | 514/40,000 = 0.01 |
| HC | $\frac{100}{160} = 0.63$ | 514/19,300 = 0.03 |
| Trace metals | | |
| Be | $10^4$ | |
| Hg | $10^2$ | |
| Pb | 50 | |

Thus, the assumption is made that an increase of 1 $\mu g/m^3$ of $SO_x$ is as detrimental as 1.25 $\mu g/m^3$ of $NO_x$ and an increase of 1 $\mu g/m^3$ of CO is as undesirable as 0.1 $mg/m^3$ of $NO_x$, etc.

A similar type of normalizing approach for the development of a combined index for the measurement of air pollution has been suggested by Babcock (ref. 2). His factors which were based upon California air quality standard are shown for comparison and have been normalized to $NO_x$.

#### *Weighting Factors for Water Parameters*

The procedure used to derive the weighting factors for the parameters within the water component is based upon the same concept as that used for the air factors, i.e., based upon water quality standards. Credence to this approach is given in the work of Brown (ref. 3) and Horton (ref. 4). In addition to these references, Wolman (ref. 5) has also identified important indicators of water quality. The water quality parameters used in the present study are:

| Importance | Weighting factor |
|---|---|
| Stable solid, leaching not important | 1 |
| Environmental pollution potential if leached or eroded | 2 |
| Hazardous if leached or eroded | 3 |
| Directly hazardous (contact, proximity etc.) | 4 |

The weighting factors were derived by normalizing to the value of dissolved oxygen.

### *Weighting Factors for Solid Waste*

Many module produce some solid waste. Consideration of the quantity of this waste also can be assessed as part of the environmental impact. The weighting factors were assigned on the basis of a gross scale of importance based on the composition of the solid waste.

### *Land-Use Parameters*

The land-use parameter includes the land area involved and a time factor. For the processing and utilization modules, the proper unit arises from the fact that a certain land area is associated with a plant with a stated throughput, as in tons of coal per hour or the equivalent heating value (Btu per hour). When the area in acres is divided by this energy rate, the resulting units are acre-hour/$10^6$ Btu. For the extraction modules, the area is associated with a total energy (e.g., tons of coal per acre). The resulting value in acres/$10^6$ Btu is converted to consistent units by multiplying the value by the length of time assumed for the operation.

Different weighting factors could be applied to reflect geographical location, compatibility with surroundings, dedicated or temporary use, and other related factors. However, weighting factors for land use have been taken as unit for all calculations performed to date.

### *Weighting Factors for Radioactivity*

A rigorous comparison of the environmental effects of material emissions and radioactive emissions from energy generation was beyond the scope of this study. One of the major difficulties is the need to consider both somatic and genetic effects for radioactivity against only somatic effects of material emissions. While lethal levels of radioactive and nonradioactive pollutants are reasonably well known, the exposures to the public encountered in energy generation are well below these lethal values. Dose-effects relationships down to the near background or natural concentrations are not known with certainty for either type of pollutant and the comparability between the assumed relationships of both types of pollutants is strictly hypothetical. Nevertheless, in this attempt to compare the environmental burden of a number of energy systems, it was desirable to develop some appreciation of magnitudes of the relative effects and to apply an internally consistent set of weighting factors to the emissions. Several approaches to derive a factor which would permit a comparison of nuclear and fossil fuel systems based upon effects were examined, and a method to include radioactive emissions under the air and water components was derived.

***Health Costs Data.*** According to the Council on Environmental Quality (ref. 6), the total annual health costs due to air pollution is \$6 billion. For a population of 200 million, the cost is \$30 per person per year.

Not all air pollution, however, is attributable to stationary sources. A simple estimate of the fraction of air pollution from stationary sources can be derived on a weight basis. The total air emissions in 1970 are (ref. 7):

Emissions, Millions of Tons Per Year

| | As Reported Fuel Combustion in Stationary Sources(a) | Total | Normalized to $NO_x$ Fuel Combustion in Stationary Sources(a) | Total |
|---|---|---|---|---|
| CO | 0.8 | 147.2 | 0.08 | 14.7 |
| Particles | 6.8 | 25.4 | 8.9 | 33.8 |
| $SO_x$ | 26.5 | 33.9 | 33.1 | 42.4 |
| HC | 0.6 | 34.7 | 0.4 | 21.8 |
| $NO_x$ | 10.0 | 22.7 | 10.0 | 22.7 |
| | 44.7 | 263.9 | 52.48 | 135.4 |

(a) Although stationary sources include more than the energy generation systems under consideration, there is little to be gained in this order-of-magnitude estimate of weighting factors to further subdivide the emissions.

The weight fraction due to stationary sources is (44.7)/(263.9) = 0.17 if the as reported data are used, and is (52.5)/(135.4) = 0.39 if the data are normalized to the $NO_x$ values.

By applying the weight fraction data to the health costs, the annual costs are $5 to $12 per person due to energy generation (stationary sources). The electric utilities used 13,750 X $10^{12}$ Btu of fossil fuel in 1970 (ref. 8). Annual health costs per person for the burning of fossil fuels to produce electricity are between $0.36 to $0.87 X $10^{-9}$ per $10^6$ Btu.

Sagan (ref. 9) has analyzed the risk to the U.S. population from all steps of the nuclear fuel cycle-occupational exposures in uranium mining and milling, manufacturing, reactor operation, fuel reprocessing, and exposure to the public near an operating reactor. His cost data are based upon the assumption that 1 rem produces 100 cases of cancer per million persons exposed and that the cost per human life is $300,000. This places the risk-cost per rem per person at $30. The annual cost for all activities is $0.11 per person.

For 10,300 megawatts of nuclear power produced by plants now operating in the United States, the human costs for the entire U.S. population of generating that electric power are derived to be about 0.026 mill per kilowatt hour. This is equal to $1.1 X $10^{-11}$ per $10^6$ Btu per person.

If health and human economic costs are used as the basis of comparison of emissions from nuclear systems and fossil systems, then emissions from the fossil fuel systems are about 100 to 300 times greater in importance than emissions from nuclear systems.

*Lethality Data.* For accidental or occupational exposures to $NO_2$, death within 3 to 5 weeks following exposure from broncheolites fibrosis obliterons results from concentrations in the range of 282 to 376 mg per $m^3$ (ref. 10). The primary ambient $NO_x$ standard is 100 micrograms per $m^3$ on an annual basis. If a breathing rate of 20 $m^3$ per day is used, the annual dose from an exposure to $NO_2$ at the allowed maximum ambient level is 730 mg/yr. If a midrange lethal exposure value of 327 mg per $m^3$ is selected, then the ratio of an instantaneous lethal concentration to allowable ambient concentrations is about 0.4.

For acute exposure to radiation, a value of 300 to 600 rads is considered lethal (ref. 11). The maximum acceptable permissible dose to the general population from manmade sources is 170 mrem on an annual basis. If a midrange lethal dose of 450 rads is selected, then the ratio of lethal dose to allowable ambient doses is about 2,600.

Thus, if the assumptions are made that the ambient or general population exposure standards are set to limit health effects, that the effects they are set to limit are equally severe, and that dose and dose-rate relationships can be extrapolated to ambient levels with the same degree of comparability, then, using this lethality approach, fossil-fuel emissions are about 6500 times more important than emissions from nuclear systems. The validity of these assumptions is not known.

*Comparison of Nuclear and Oil-Fired Power Plants.* An analysis has been performed by Starr et al. (ref. 12) to compare emissions from nuclear and oil-fired power plants in the Los Angeles basin. The analysis was restricted to the comparative public health aspects of oil-fired and nuclear power plants and their associated activities in a typical urban setting. Operation of these plants, under conditions up to the present Federal regulatory limits, was estimated to cause 60 times more respiratory deaths due to fossil fuel pollution than cancer deaths due to nuclear plant effluents. In normal practice, neither plant would be expected to expose the public to these limits, primarily because the routine effluents must be reduced below regulatory levels to meet a variety of conditions, and would thus be expected to be substantially less (by a factor of 10 or more) under average circumstances.

In both cases the integrated accident risk (averaged over time and all episodic events) is about $10^{-5}$ of the continuous exposure, for either the nuclear plant or the oil-fired plant. For the analyzed accident with equal probability of occurrence, the oil-fired plant has a substantially worse public health impact than the nuclear. For example, the one-in-a-million-years event for the oil-fired plant would lead to approximately 700 respiratory deaths in a population center of 10 million people; while the one-in-a-million-years event for the nuclear plant would result in approximately one death in the same population.

Data from Los Angeles are not directly applicable to other regions of the country. However, the analysis by Starr is useful in establishing the relative significance of nuclear power and fossil fuel power emissions. The factor of 60 which he derived is of the same order of magnitude as the factor of 100 derived from health cost effect.

*Derivation of Weighting Factor ($W_p$) for Radioactivity.* In order to compare the emissions from nuclear power systems (derived in units of curies) to the emissions from fossil fuel systems (derived in units of pounds), it was necessary to develop a conversion factor which both converts the units to the same basis and relates comparable effects. The previous discussion of health costs and lethality data gives an estimate of the relative importance of exposure to fossil fuel emissions and to radioactivity. It would appear that fossil fuel emissions are at least 100 times more important than nuclear emissions on an effect basis. In light of the uncertainties in the analysis and the limited consideration of accidents in nuclear power systems, it has been assumed that emissions from fossil fuel systems and emissions from nuclear systems have equal importance. For normal operations, this will tend to overstate the importance of the environmental impact of nuclear power.

Comparison of emissions from the two types of systems would require a detailed analysis of each specific pollutant or radionuclide emitted. For the purposes of this study, several approximations were made. During reactor operations and fuel reprocessing, it is assumed that the air emissions are predominantly the noble gases. MPC values (10 CFR, Part 20) for the noble gases are $3 \times 10^{-7}$ $\mu$Ci/ml. This value will be taken as the standard for the air emissions calculations. Several radionuclides can be present in the waste water, and MPC values (10 CFR, Part 20) in water for some typical radionuclides are: $^{3}$H, $3 \times 10^{-3}$ $\mu$Ci/ml; $^{137}$Cs, $2 \times 10^{-5}$ $\mu$Ci/ml; and $^{90}$Sr, $3 \times 10^{-7}$ $\mu$Ci/ml. For purposes of the water effluent calculations, a midrange value of $3 \times 10^{-5}$ $\mu$Ci/ml has been used for the "standard" MPC water value.

To reduce the radioactivity to the same units as the fossil fuel emissions, the following relationships are applied.

***Air emissions.***

$$\text{Radioactivity in NO}_x\text{ equivalents} = Q_{r,\text{air}} \times \frac{I_{\text{NO}_x}}{I_r} \times \frac{\text{Ambient standard NO}_x}{\text{``Standard'' MPC air}}$$

where $Q_r$ = quantity of radioactivity in curies

$\dfrac{I_{\text{NO}_x}}{I_r}$ = Factors which relates the importance of fossil fuel emissions to radioactivity in units of lb/Ci. For this analysis it is assumed to be 1.

$$\text{Ambient standard, NO}_x = 100\ \mu\text{g/m}^3 = 0.23 \times 10^{-6}\ \text{lb/m}^3$$

$$\text{``Standard'' (MPC) air} = 3 \times 10^{-7}\ \text{Ci/m}^3$$

$$\text{Radioactivity in NO}_x\text{ equivalents} = Qr \times 0.7,\ \text{lb.}$$

*Water effluents.* To derive a similar factor for water, radioactivity has been compared to trace metals. As noted earlier, the water quality standard values, based primarily on drinking water standards, are in the range of 0.1 to 0.3 ppm with the Hg-standard set of 0.5 ppb. For the purpose of comparison, a water quality value of 0.1 ppm will be used.

$$\text{Radioactivity in trace metal equivalents} = Q_{r_{\text{l water}}} \times \frac{I_{\text{trace metals}}}{I_r} \times \frac{\text{Water quality standard trace metals}}{\text{``Standard'' (MPC) air}}$$

As in the case of air, $I_{\text{trace metals}}/I_r$ is a factor which relates the importance of trace metals to radioactivity. It is assumed to be 1 pound per curie for this analysis.

$$\text{Water quality standard, trace metals} = 0.1\ \text{ppm} = 2.2 \times 10^{-4}\ \text{lbs/m}^3$$

$$\text{``Standard'' (MPC) water} = 3 \times 10^{6}\ \text{Ci/m}^3$$

$$\text{Radioactivity in trace metal equivalents} = Q_r \times 7,\ \text{lb}$$

Since the water impacts are normalized in terms of dissolved oxygen, an additional factor of 50 has been applied to the preceding expression. Thus, in order to calculate the burden to the water from radioactivity, the radioactive effluents in curies is to be multiplied by $3.5 \times 10^{-2}$ to obtain a comparable unit.

*Ecology*. Weighting factors for ecological impacts to energy systems have not been estimated for the current study. Concepts to be used, however, could follow the general approach outlined in the study for the Bureau of Reclamation (ref. 1). The ecological impacts are not included in the current environmental index.

*Human Factors*. A combination of parameters is involved in the impact of energy systems directly upon man's environment. Data needed to derive quantitative weighting factors are often missing, and only a subjective approach is possible at this time. For this report, only the occupational health and safety sections have been considered. These have been derived primarily of data from the Bureau of Labor Statistics (ref. 13). Weighting factors have not been derived, and the human factor impacts are not included in the current environmental index.

### *Computer Program for Environmental Index Calculation*

The computer program for the calculation of the environmental index for the energy systems evaluated encompasses seven steps listed below:

(1) *Read-in Data*

The data read-in consists of:

- (a) Emission values for the components air, water, solid, and land for each module;
- (b) The weighting factor for each emission value within the component;
- (c) Weighting factors for the relative importance of air, water, solid, and land;
- (d) Weighting factors for module type, extraction, transportation, processing, and utilization;
- (e) The modules comprising each system.

(2) *Calculate Weighted Component Totals for Each Module*

This calculation is performed by multiplying the emission values within each component by the appropriate weighting factor (read in 1-b). The result is four numbers per module representing the sum of impacts for each component, air, water, solid, and land.

(3) *Efficiency Correction*

The weighted component totals for each module are divided by the product of the efficiencies of all subsequent modules in the system. This calculation is necessary since the module data are expressed on a basis of a million Btu. The output, and thus the associated emissions, of, say, an extraction module, must be increased in proportion to the inefficiency of the power plant.

(4) *Calculate Normalizing Factors*

The modules are arranged into systems (according to data read in 1-e), and the air, water, solid, and land totals are summed up. For the number of systems chosen, this procedure results in a total of four sums: air, water, solid, and land. The normalizing factor is then calculated as the ratio of the air sum to each of these sums.

(5) *Calculate the Module Index*

For each module in a system, the four normalized numbers (air, water, solid, land) are weighted with respect to component weighting factors (read in 1-c) and summed. This calculation results in a single impact number for each module in a given system.

(6) *Calculate the System Index*

For each system, consisting of several modules, the system index is calculated by multiplying each module impact number by the module weighting factor (read in 1-d). The sum of these results within a given system gives the system index.

(7) *Rank the System*

Finally, ranking of the systems is performed by ordering by system index.

### *Sensitivity Analyses*

The procedures used in deriving an environmental index for various energy systems incorporated a number of assumptions; in many cases, estimates have been made of the quantities of pollutants emitted in the modules. A series of analyses were performed, using the computer code previously described, to test the sensitivity of the ranking of the systems to variations in the input and the assumptions that have been made. The results of these sensitivity analyses given an indication of the validity of the overall procedure and an appreciation of the reliability of the final ranking.

The results of three sets of calculations are shown in table A-I to illustrate the changes in rank ordering of systems that occur with changes in weighting factors. The base case for comparison is the ranking systems where all components and modules were equally weighted. These rankings are compared to a case where the solid waste component was weighted at 30 percent of the other component weighting factors; i.e., burdens as a result of solid wastes are less important than burdens in air, water, and land utilization components. In the third case, all of the components were equally weighted, but the utilization module was weighted 3 times as great as the other modules.

In general, the overall rankings remained similar; the systems that rank high in the base case tend to rank high in the comparison case. Where the importance of solid waste component is diminished, the system using Eastern coal and a limestone

Table A-1. Sensitivity of Ranking to Variations in Weighting Factors

| All Components and Modules Weighted Equally | $W_A = W_W = W_L = 1.00$; $W_{Solid} = 0.3$ All Modules Weighted Equally | All Components Weighted Equally Utilization Modules Weighted 3 Times Greater Than Others |
|---|---|---|
| LNG | LNG | LNG |
| Fluidized bed-oil | Fluidized bed-oil | Fluidized bed-oil |
| Gas | E coal-molten iron | Import oil-topping-CB |
| E coal-molten iron | E coal-pres fluid bed | Gas |
| W coal-conv boil | N gas-GP-CB | E coal-SRC-CB |
| E coal-pres fluid bed | W coal-CB+LSS | E coal-molten iron |
| E coal-MgO scrubber | E coal-CB+MgO | Off coi-pipe-ref-OT-CB |
| Import oil-topping-CB | E coal-PC-CB+LSS | E coal-pres fluid bed |
| E coal-PC-CB | W coal-CB | E coal-PC-CB+LSS |
| E coal-PC-CB+LSS | E coal-CB+LSS | On oil-pipe-ref-barge-CB |
| W coal-CB+LSS | E coal-SRC-CB | On oil-pipe-crude gas-pipe-gas CB |
| On oil-pipe-ref-barge-CB | E coal-PC-CB | W coal-CB |
| SRC-CB | Imp oil-topping-CB | E coal-PC-CB |
| CCC-CB | E coal-CCC-CB | E coal-MgO scrubber |
| On oil-oil pipe-crude gas-gas pipe-gas CB | E coal-Lurgi | CCC-CB |
| E coal-Lurgi | On oil-pipe-ref-barge-CB | W coal-CB=LSS |
| E coal-CB+LSS | On oil-pipe-crude gas-pipe-CB | E coal-CB+LSS |
| Off oil-oil pipe-ref-OT-CB | E coal-CB | E coal-Lurgi |
| E coal-CB | DMEC-PC-CB+LSS | On oil-pipe-ref-OT-CB |
| On oil-oil pipe-ref-OT-CB | Off oil-pipe-ref-OT-CB | DMEC-PC-CB |
| DMEC-PC-CB | DMEC-PC-CB | DMEC-PC-CB+LSS |
| DMEC-PC-CB+LSS | On oil-pipe-ref-OT-CB | E coal-CB |
| DMEC-CB | DMEC-CB | DMEC-CB |

scrubber ranks considerably higher than in the base case. The large quantities of solid waste produced in this system have relatively less influence on the ranking of this system. The imported oil system moves high in the ranking when greater weight is given to the utilization module. Its utilization module has a relatively low burden where compared to other systems. The changes in the order of the systems are in the direction anticipated by the changes in weighting of components or models.

The sensitivity analyses performed to date are not sufficient to determine the significance of small differences in environmental indices. Until additional analyses are performed and the results evaluated, care must be taken in the interpretation of the ranking and the values assigned to the environmental index.

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### *Acknowledgment*

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# THE ENVIRONMENTAL IMPACT OF COAL-BASED ADVANCED POWER GENERATING SYSTEMS

**Fred L. Robson and**
**Albert J. Giramonti***

## INTRODUCTION

The electric utility industry in the United States is currently faced with a multitude of problems. The near-term necessity of meeting promulgated environmental regulations has been exacerbated by the shortage of low-sulfur fuels and, where available, by their very high costs. A combination of events culminated in a very tight money market at the very time that utilities had to raise large sums to finance the more costly conventional and nuclear stations now being ordered. Thus, it is apparent that there exists a pressing need for an efficient, low-cost power generating system that could utilize high-sulfur fuel.

One such system that has the potential of generating power efficiently while using less costly equipment is the COmbined Gas And Steam (COGAS) power system. This advanced power generating scheme, shown in figure 1 in its simplest form, has a gas turbine exhausting into a waste heat recovery boiler which raises steam for use in a steam turbogenerator. Combined-cycle systems, as with other advanced technology gas turbine-based power systems, require fuels of utmost cleanliness; cleanliness that is usually well beyond that required by environmental regulations.

When considering the use of coal as fuel for such power systems, it is apparent that considerable processing and cleanup must precede its introduction into the engine. The following paper discusses the factors affecting the performance of combined-cycle power systems and the compromises in the system that result when this system is integrated with a coal gasifier and fuel gas cleanup process.

## PERFORMANCE OF COGAS SYSTEMS

The performance of the simple waste-heat recovery-type COGAS system shown in figure 1 can be approximated by:

$$\eta_c = \eta_{gt} + (1-\eta_{gt})\,\eta_s \left(\frac{T_{ex} - T_s}{T_{ex} - T_c}\right) \qquad (1)$$

where

$\eta_c$ = COGAS efficiency
$\eta_{gt}$ = gas turbine efficiency
$\eta_s$ = steam cycle efficiency
$T_{ex}$ = temperature of gas turbine exhaust
$T_s$ = temperature of stack
$T_c$ = temperature of ambient conditions.

The most important factor in the above equation is $\eta_{gt}$, the efficiency of the gas turbine.

### *Gas Turbine Performance*

The key to gas turbine performance is turbine inlet temperature. Currently, the newer turbines being used in utility applications operate at temperatures ranging from 1,800°F to near 2,000°F, with efficiencies of 28 percent to over 35 percent. A second significant parameter, the pressure ratio (compressor discharge pressure/ambient pressure), varies from around 8:1 to nearly 15:1 in these machines. The relationship between efficiency, inlet temperature, and the pressure ratio is shown in figure 2. The parameter specific power in this figure is a measure of the amount of work which can be done by a given size machine and is, indirectly, a guide to cost, i.e., a machine with a high specific power will generally have a lower cost in $/kW than a machine of lower specific power. Prior studies (refs. 1,2) indicate that turbine inlet temperatures over 2,000°F could be expected during the 1970 decade.

### *Steam System*

Although the efficiency term for the steam cycle in eq. 1 is modified by several coefficients, it is apparent that it is desirable to achieve as high an efficiency as practicable. There are, however, constraints on the steam cycle conditions which are external to the steam power system. These are gas temperature to the boiler (gas turbine exit temperature) and the stack temperature. The latter is set by dew point

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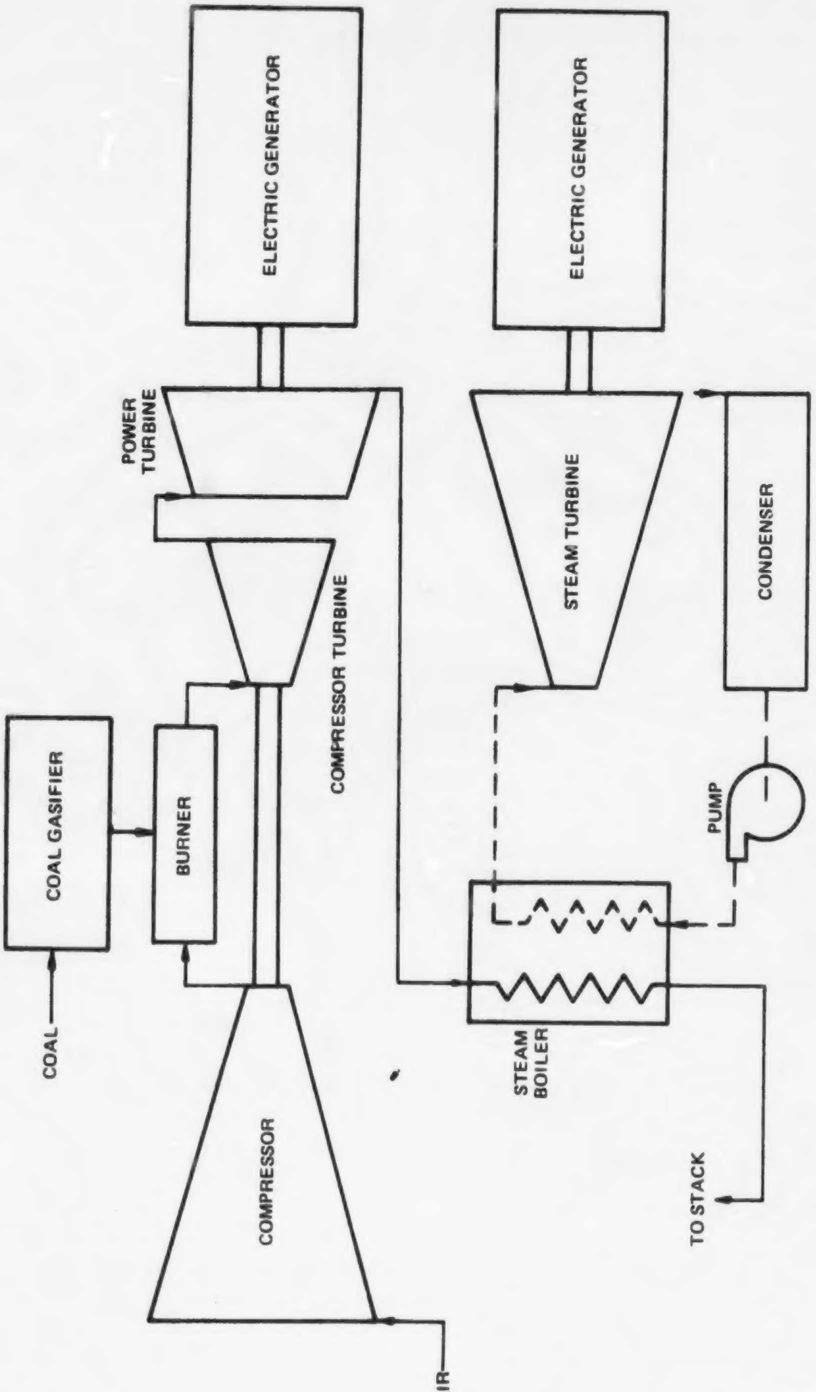
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ELECTRIC GENERATOR
POWER TURBINE
STEAM TURBINE
CONDENSER
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COAL
STEAM BOILER
COMPRESSOR
TO STACK
AIR

Figure 1. Combined Gas-Steam Turbine System

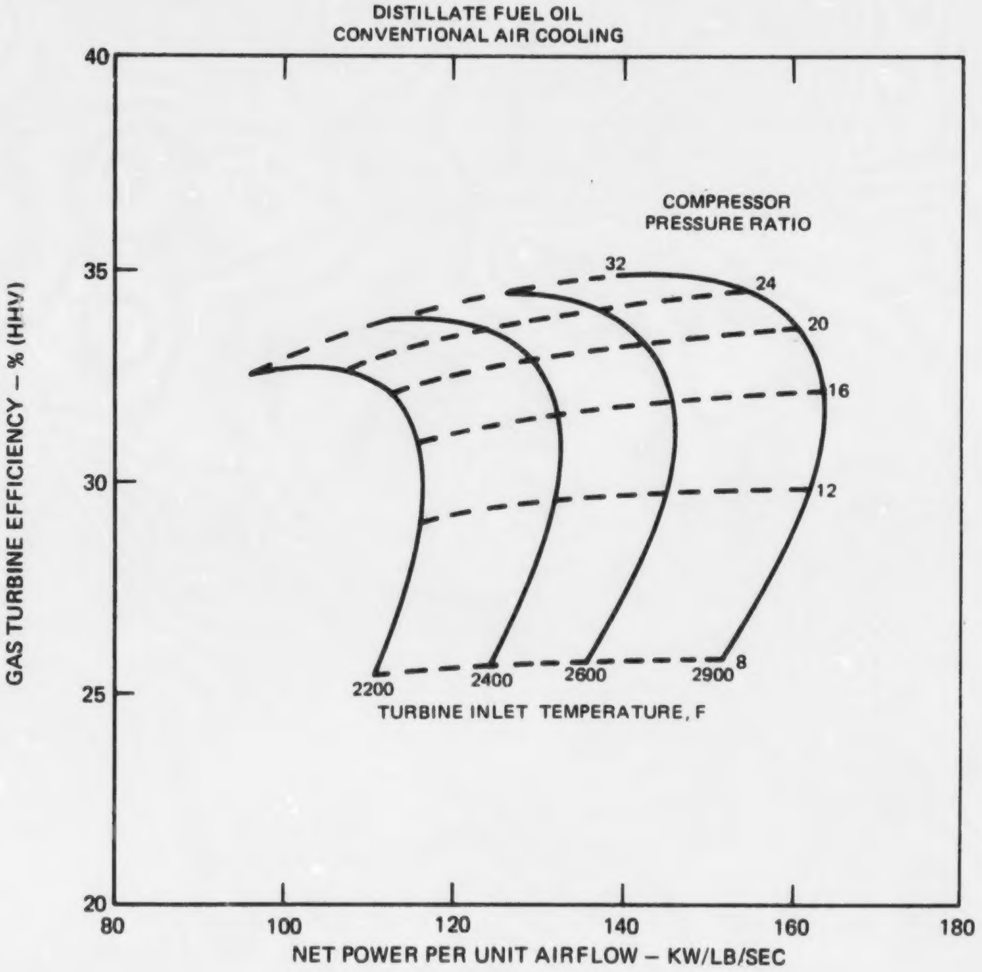


**Figure 2. Gas Turbine Performance**

considerations and is usually not much below 300°F, even when a clean fuel is used. It has been found (ref. 2) that a pinch temperature difference of 100°F results in the most acceptable heat exchanger design. Thus, for systems utilizing waste-heat recovery boilers, the maximum steam temperature is 100°F less than the gas turbine exhaust temperature.

For COGAS systems operating with conventional distillate or natural gas fuels, the most attractive steam system would have a quite simple cycle configuration. Regenerative feedwater heating would be limited to a single deaerating feedwater heater. The generation of steam would be at a single pressure level, and essentially all the steam would pass through the turbogenerator. As will be discussed in a subsequent section, the presence of a coal gasification system alters the makeup of the steam system.

*COGAS Performance*

The potential performance of the COGAS system is one of its most attractive attributes. When referring to a system using conventional distillate-type fuels, performance levels as shown in figure 3 could be attained. At 2,200°F turbine inlet temperature, a level which should be attainable in base-load turbines in the mid-to-late 1970's, an efficiency of 43.5 percent could be realized. As turbine inlet

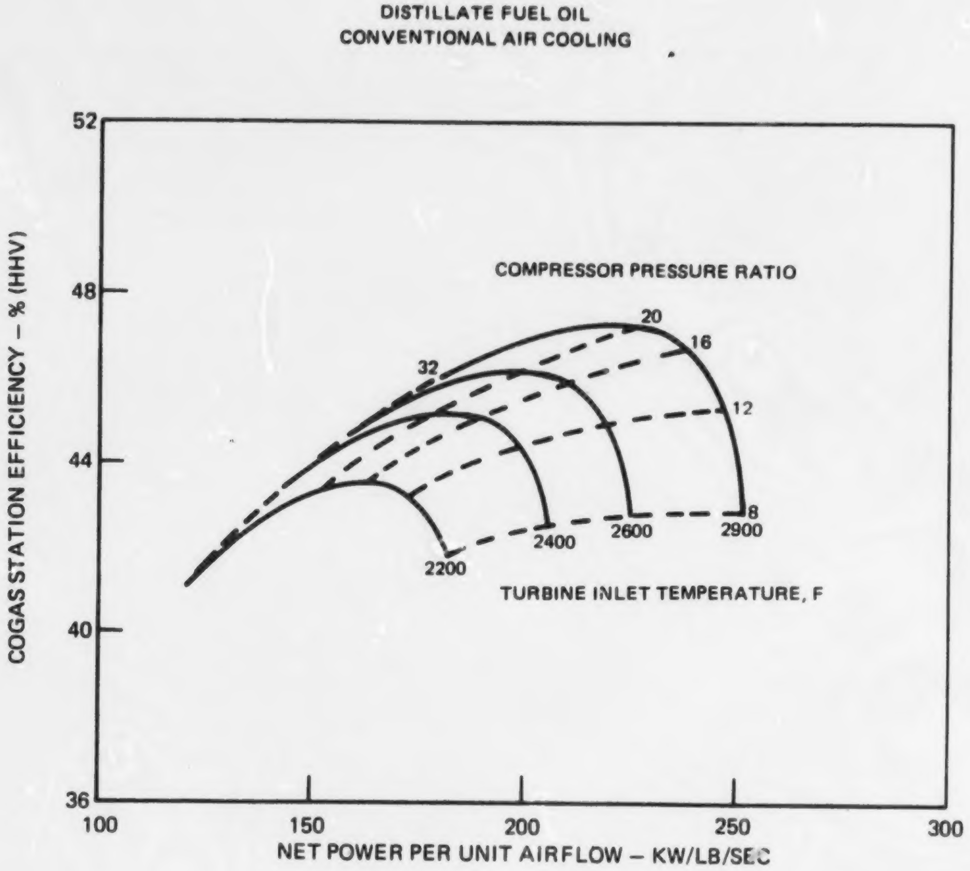


**Figure 3. COGAS Station Performance**

temperatures increase, the efficiencies also increase, and it should be possible to approach 50 percent efficiency at inlet temperatures of approximately 3,000°F. The values in figures 2 and 3 are based on conventional turbine cooling techniques. The use of advanced cooling techniques or advanced materials could significantly increase these values. Results in reference 3 indicate COGAS efficiencies of about 55 percent at 2,900°F.

Figure 3 also shows the effect of pressure ratio on performance. It is interesting to note that the COGAS system efficiency is not particularly sensitive to pressure ratio, whereas specific power is sensitive to pressure ratio. Therefore, tradeoffs between high efficiency and high specific power can be considered without undue performance penalty.

COGAS efficiency is replotted in figure 4, this time against the parameter fraction of the fuel utilized in the gas turbine. As the fraction of fuel utilized in the gas turbine decreases (more fuel burned in boiler to raise steam), the efficiency also tends to decrease. The steam conditions in figure 4 have been adjusted to take into account the additional firing. Thus, the steam cycle efficiency is increasing as more fuel is burned. At the lower turbine inlet temperature, the point of maximum efficiency occurs with very slight firing, but the small increase in efficiency would be more than offset by the additional cost of the more efficient steam system. At the higher turbine inlet temperatures, the point of maximum efficiency occurs when steam is being raised by exhaust heat alone and all the heat added to the cycle is used at combined-cycle efficiency. As more steam is generated, additional fuel must be burned. This

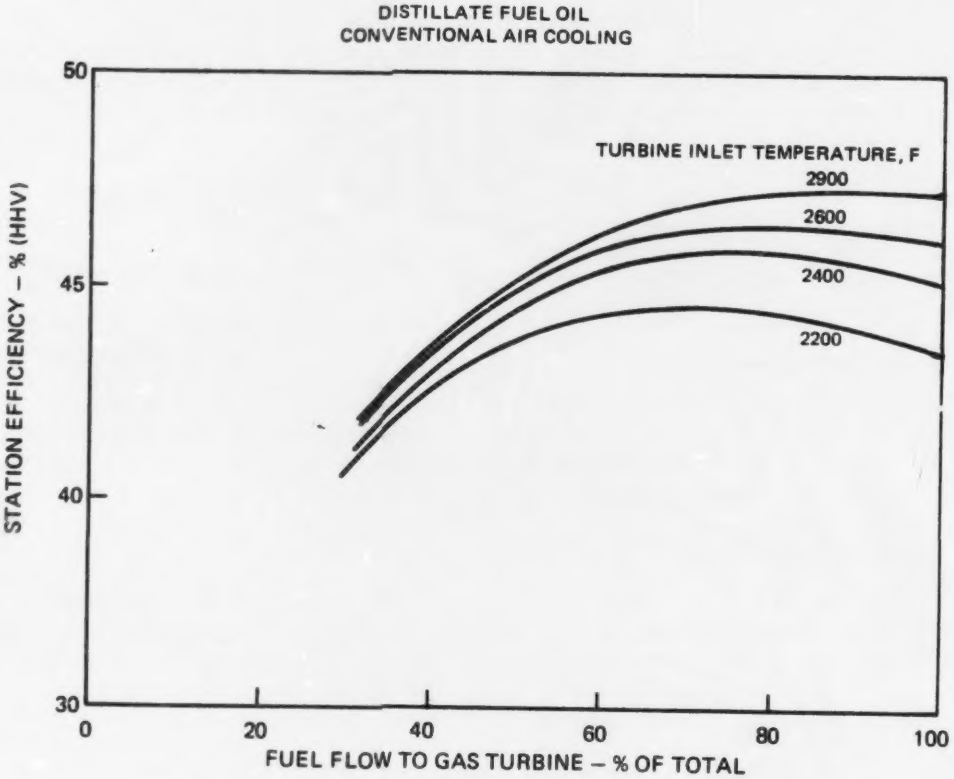


**Figure 4. COGAS Station Performance With Fired Boiler**

additional fuel increment is used only at steam-cycle efficiency resulting in a degradation of the overall system efficiency. This point is quite important in understanding the overall performance of integrated gasification, cleanup process, and COGAS systems.

Having established some base-line performance estimates for COGAS systems using conventional fuels, let us examine some representative processes that produce low-Btu gas from coal and also some processes that clean the low-Btu gas to meet standards for both environmental and engine requirements.

## COAL GASIFICATION PROCESSES

Production of low-Btu fuel gas from coal is achieved by gasification with air and steam at elevated temperatures. The overall gasification process is endothermic, primarily due to the steam-carbon reaction which requires about 5,000 Btu per pound of carbon:

$$C + H_2O \rightarrow CO + H_2 \qquad (2)$$

This heat requirement is satisfied by burning a portion of the coal feed with air.

Gasification systems are commonly classified into three categories according to the characteristics of the coal bed, i.e., fixed, fluidized, or entrained. Generally speaking, fixed-bed systems operate with countercurrent flow of coal and gas at temperatures below the ash-fusion point. Consequently, these systems are characterized by relatively low gasification rates and substantial tar formation. High-temperature gasification under ash slagging conditions affords higher gasification rates and provides essentially a tar-free producer gas. Cocurrent, entrained-flow gasifiers typically operate under the latter conditions. However, at higher temperatures, the fraction of the coal heating value represented by sensible heat of the product gas is substantially greater than for fixed-bed gasifiers, and either high-temperature cleanup systems or heat recovery systems must be employed to achieve comparable thermal efficiencies.

For use in advanced-cycle power systems, the product gas must be at an elevated pressure. Since the volume of the product gas is roughly twice that of the air required for gasification, it is generally advantageous to employ pressure gasification rather than product gas compression to obtain a given delivery pressure. Moreover, the specific gasification rate (gas produced per unit of reactor volume) is greater at higher pressure.

### *Gasification System Characteristics*

Present-day technology for pressure gasification of coal is limited to the fixed-bed type gasifier. Second generation technology, judged to become available in the 1980 decade, will probably evolve from current developmental efforts on entrained-flow and fluid-bed gasifiers. In the current study being carried out by UARL and Foster Wheeler, the BCR two-stage, entrained-flow-type gasifier was selected as representing advanced coal gasification technology. Typical operating characteristics of fixed-bed and entrained-flow gasifiers are given in table 1.

The prominent differences in the performance of these gasifiers are directly related to operating temperature:

(a) The relatively low carbon conversion for the fixed-bed system and the formation of byproduct tar;

(b) The ratio of sensible heat of product gas to coal heating value is twice as great for the entrained-flow gasifier.

In the case of the fixed-bed gasifier, the presence of tarry material in the gas complicates downstream heat recovery and subsequent gas purification. For low-temperature purification of the raw gas, direct quenching is the preferred method thereby degrading the level for sensible heat recovery. However, the recovered tar product could be recycled to the gasifier, thereby increasing the overall carbon gasification. If a high-temperature purification system is integrated with the fixed-bed gasifier such that no tar is allowed to condense, then the tar represents a component in the delivered product gas. This may present a problem from a pollution standpoint in that the tarry material typically contains 1.5 to 3.0 percent sulfur and about 1.0 percent nitrogen, most of which would convert to $SO_2$ and $NO_x$ during combustion.

The large sensible-heat content of the gas from an entrained-flow gasifier makes it imperative, from an efficiency standpoint, to use heat recovery techniques when used in conjunction with low-temperature purification. Fortunately, the absence of condensible tars facilitates indirect heat exchange and affords the possibility for reheating the fuel gas feed to a gas turbine. Because of the high effluent temperature, the entrained-flow gasifier is particularly amenable for integration with high-temperature purification systems operating in the range of 1,600° to 1,700°F.

## FUEL GAS CLEANUP PROCESSES

The fuel gas leaving the gasification system contains sulfur- and nitrogen-bearing compounds and particulates which must be removed to meet constraints set by environmental concern and by gas turbine requirements. The extent of the latter requirements are noted in table 2 where the fuel contaminant constraints are compared to values of contaminants in the raw fuel gas. The necessity for some sort of cleanup is apparent.

Cleanup systems can be categorized in many ways; however, when mated with gasification and power systems, it is useful to consider processes as operating either at a low temperature or at a high temperature. The dividing line between the two is somewhat arbitrary but, for convenience, can be considered as 250°F.

### *Low-Temperature Cleanup Process Characteristics*

Low-temperature cleanup systems for desulfurizing raw producer gas are commercially available and have been widely used for natural gas sweetening and in treating synthesis gas for petrochemicals, e.g., ammonia, methanol, and oxo-alcohols. Commonly, these are liquid scrubbing systems which operate below 250°F. They are usually classified according to the mode of acid gas absorption by the solvent, i.e., either physical or chemical absorption. Table 3 lists several typical low-temperature sulfur removal processes, together with the type of absorbent and approximate operating temperatures.

Physical solvent processes generally employ an organic solvent which absorbs the acid gas components in accordance with their partial pressures in the gas phase. Chemical systems usually employ aqueous solutions of inorganic or organic compounds which chemically react with the acid gas components. These systems are relatively insensitive to partial pressure effects. In both types of systems, the absorbent can be regenerated either by heat, pressure reduction, or a combination of these, thereby producing a gas stream rich in sour gas components.

Table 1. Operating characteristics for two representative gasification processes

| Gasifier Type: | Fixed-Bed | Two-Stage Entrained-Flow |
|---|---|---|
| Temperature, F | 1000 | 1800 |
| Pressure, psia | 500 | 500 |
| Coal | Western Kentucky | Illinois No. 6 |
| Air/Coal, lb/lb | 2.69 | 3.42 |
| Steam/Coal, lb/lb | 0.349 | 0.567 |
| % Carbon Gasified* | 84 | 99 |
| Producer Gas | | |
| Vol. % | | |
| $N_2$ | 47.61 | 47.70 |
| CO | 20.55 | 16.74 |
| $CO_2$ | 5.88 | 8.84 |
| $H_2$ | 13.83 | 11.98 |
| $CH_4$ | 2.76 | 3.14 |
| $H_2S$ | 0.60 | 0.46 |
| COS | 0.10 | 0.10 |
| $NH_3$ | 0.25 | 0.38 |
| $H_2O$ | 8.42 | 10.66 |
| | 100.00 | 100.00 |
| Tar, lb/lb coal | 0.11 | 0.0 |
| HHV, Btu/SCF (Tar Free) | 139 | 125 |
| Sensible Heat/Coal HHV, % | 12.2 | 24.6 |

*No tar recycle

## Table 2. Coal-based gaseous fuels

| Impurity | Concentration | Max Allowable |
|---|---|---|
| Solids | > 0.1 g /$ft^3$ | 0.8 lb/$10^6$ $ft^3$ |
| Condensable Hydrocarbons | 0-100 lb/$10^6$ $ft^3$ | 0.5 lb/$10^6$ $ft^3$ |
| Sulfur | Variable | 0.18% Mol ($H_2S$) |
| Metals | | |
| Vn | 10 ppm | 0.1-0.2 ppm wt |
| Na + K | NA | 0.2-0.6 ppm wt |
| Ca | NA | 0.1 ppm wt |
| Pb | 3 ppm | 0.1 ppm wt |
| Cn | NA | 0.02 ppm wt |

## Table 3. Representative low-temperature sulfur removal processes

| Type | Absorbent | Temp., F | Status |
|---|---|---|---|
| Chemical | Aqueous $K_2CO_3$ | 160-230 | Commercial |
| Chemical | Potassium Di-Methyl Amino Acetate | 85 | Commercial |
| Chemical | Alkanol Amine | 100 | Commercial |
| Chemical & Physical | Sulfolane + Di-Isopropanolamine | 120 | Commercial |
| Physical | Polyethylene Glycol Ether | 50 | Commercial |
| Physical | Methanol | -40 | Commercial |
| Physical | N-Methyl Pyrrolidone | 80 | Commercial |

Selection of those systems most attractive for treating coal-derived fuel gas must take into consideration the following:

1. Sulfur removal capabilities, not only with respect to $H_2S$, but also other sulfur compounds such as COS and $CS_2$;
2. Selective absorption of sulfur compounds over carbon dioxide; since $CO_2$ need not be removed from fuel gas intended for use in advanced power cycles, the $CO_2$ absorption represents an increased operating load on the system in terms of solvent circulation, regenerative heat requirements, and equipment sizes;
3. Type of absorbent insofar as the purified gas may be contaminated by entrained or volatilized solvent;
4. The system tolerance to trace contaminants in the raw producer gas such as HCN, phenols, tars;
5. Overall energy requirements and capital cost.

Generally speaking, liquid scrubbing systems are capable of reducing the sulfur content in the treated gas to 100 ppm or less. These systems are also capable of removing the particulates from the fuel gas stream to levels compatible with the gas turbine requirements.

### *High-Temperature Cleanup Process Characteristics*

High-temperature systems for sulfur removal are not commercially available, although there are several in various stages of development. Most of the active work presently involves use of limestone and dolomite, which have potential in the range of 1,500° to 2,000°F. Other systems receiving attention employ iron oxides, molten salts, and liquid metals. To date, none of these systems have been demonstrated on the pilot plant level.

Table 4 lists several processes currently under development which may prove viable for use with second generation gasification systems. These systems operate by chemical reaction of the absorbent with sulfur compounds in the gas yielding the corresponding metal sulfides. As with low-temperature systems, economics require that the spent absorbent be regenerated for reuse. Depending upon the chemical system, regeneration is conducted with either air or steam plus carbon dioxide, e.g.:

$$CaS \cdot MgO + H_2O + CO_2 \rightarrow CaCO_3 \cdot MgO + H_2S;$$
$$2\ FeS + 3\tfrac{1}{2}O_2 \rightarrow 2\ SO_2 + Fe_2O_3.$$

Selection of potentially attractive high-temperature systems should consider the following factors:

1. Capability for removing sulfur compounds, COS as well as $H_2S$. The residual concentration of sulfur in the treated gas is determined by chemical equilibria for each system. In general, high-temperature systems do not appear to be as versatile as wet scrubbing systems in terms of reducing residual sulfur to 100 ppm or less.
2. The form in which absorbed sulfur is

Table 4. Representative high-temperature sulfur removal processes

| Process-Type | Absorbent | Temp. F | Status |
|---|---|---|---|
| Half-calcined dolomite | $CaCO_3 \cdot MgO$ | 1500-1800 | Pilot |
| Fully-calcined dolomite | $CaO \cdot MgO$ | 1600-2000 | Pilot |
| Sintered iron oxide | $Fe_2O_3$ + Fly Ash | 800-1500 | Pilot |
| Iron oxide | $Fe_2O_3$ | 700-1200 | Experimental |
| Molten carbonate salt | $Na_2CO_3 + C_aCO_3$ | 1100-1700 | Pilot |

regenerated, $H_2S$, $SO_2$, or elemental sulfur. Elemental sulfur is the preferred form, since it can be stockpiled without presenting significant pollution problems.

3. Ability to regenerate the absorbent without substantial loss in activity.
4. Overall energy requirements and projected capital costs.

At this time, it is not apparent that the high-temperature cleanup processes would remove particulates effectively. Additional particulate removal devices might have to be used, but such devices have not yet been identified.

## PERFORMANCE OF INTEGRATED SYSTEMS

The characteristics of the three major systems which make up the integrated powerplant have been described. The combinations of these systems give rise to a very large number of possible configurations, especially when the necessary auxiliaries such as heat exchangers, booster compressors, etc., are taken into account. In order to investigate these many combinations, a computer model was developed at UARL under corporate sponsorship which allows great flexibility in analyzing the integrated systems. The basic model is shown schematically in figure 5; in actual use, there are many more modules.

The three columns in figure 5 represent the COGAS system, the gasifier and cleanup stream, and an air preheater fired by the product gas. This latter equipment is but one of several ways of preheating air before its use in the gasifier.

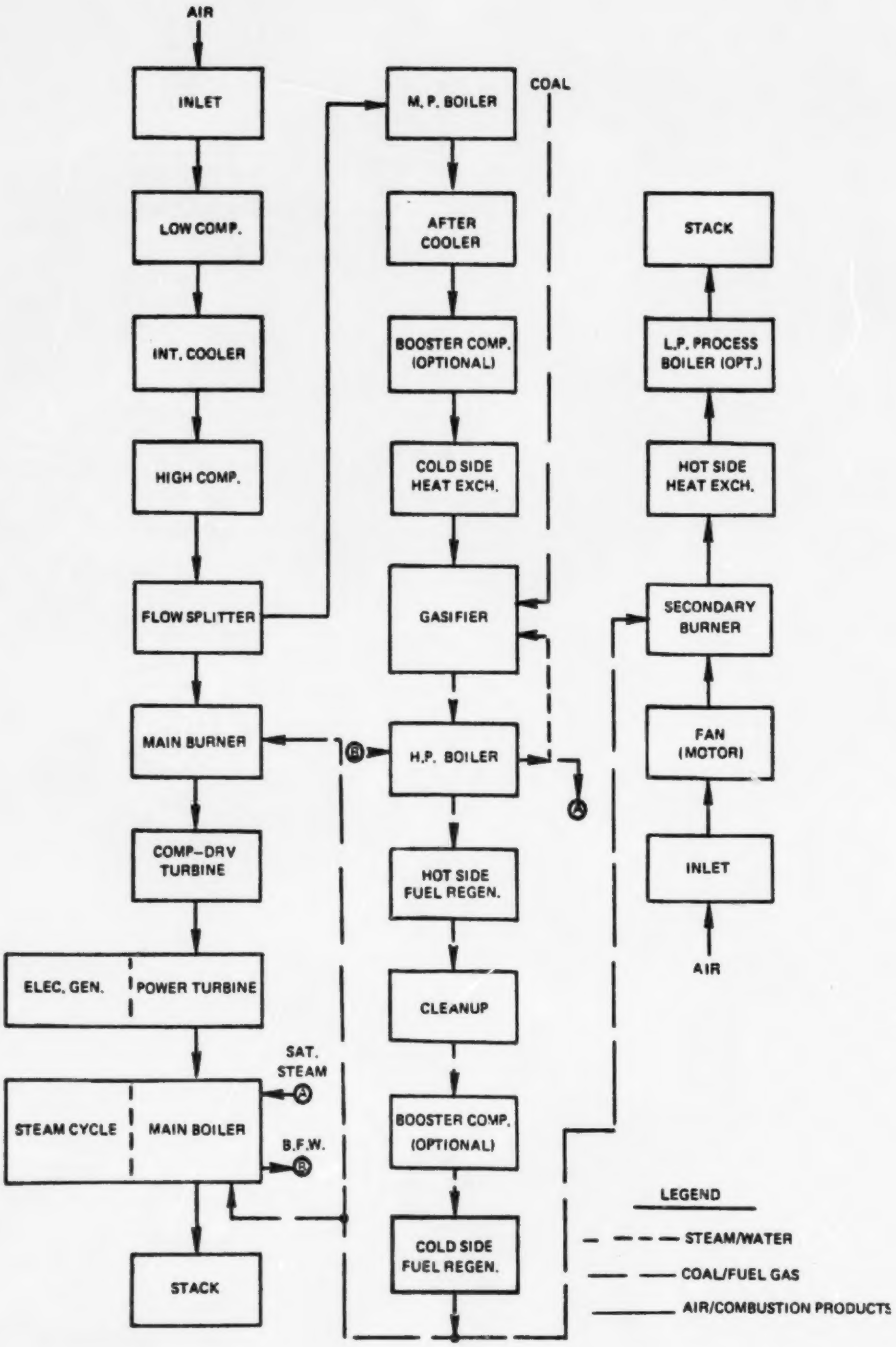
While the following is applicable to nearly all gasification processes, for the purpose of this discussion, only the high-temperature entrained-flow process will be considered.

In the integrated power system, air for the gasifier is bled from the gas turbine compressor. In order to overcome the various pressure drops in the gasifier, cleanup, and gas turbine fuel controls, the gasifier must operate at a pressure above that of the gas turbine; thus, a booster compressor is required. One method of driving this compressor is by use of a steam turbine using high-pressure steam from the main steam boiler (see fig. 6 for a simplified flow chart of one of the many possible configurations). After gasification, the hot fuel gas must be purified. If a high-temperature method were used, the only box appearing after the gasifier in figure 5 would be that for the cleanup system. If a low-temperature system were used, the hot fuel gas would pass through one or more waste-heat boilers to drop its temperature from roughly 1,750°F to a level acceptable for the cleanup system. Alternatively, the hot gas could be cooled by regeneration against the cooler, purified fuel gas; or, a combination of the two could be used. Yet another alternative is shown in figure 6 where a waste heat boiler is used in conjunction with a heat exchanger in which the cool, purified fuel gas is heated against the hot bleed air from the compressor prior to recompression in the booster compressor. The attractiveness of such a system is somewhat doubtful, however, because leakage in this heat exchanger could produce a potential explosion hazard.

The reasoning behind all of these schemes is to recover the sensible heat in the fuel gas, which, in the two-stage entrained-flow-type gasifier, can be as much as 25 percent of the total fuel heating value. With high-temperature cleanup, nearly all the sensible heat would be used in the gas turbine combustion at combined-cycle efficiency, and very little steam would be raised by burning fuel. (Steam raised in the gasifier waste-heat boiler is, of course, generated by heat obtained by burning coal in the gasifier.)

In low-temperature systems, the sensible heat would be recovered in a waste-heat boiler or in a fuel gas regenerator. As was pointed out in figure 4, the generation of steam by burning fuel is generally undesirable. This may be seen best by examining figure 7, which compares distillate-fired and gasified-coal-fired COGAS system performance. In this figure, the normalized efficiency of a conventional distillate-fired COGAS system is shown as a function of the fraction of the fuel input used to raise steam. The normalized efficiency would be unity when all the fuel used is burned in the gas turbine and would be about 0.85 when 25 percent of the total fuel used is burned in the boiler (unlike fig. 4, the steam conditions remain constant in fig. 7). The values of efficiency for gasified-coal-fired systems using several different cleanup processes, for both high and low temperatures, have also been normalized against the distillate-fired system.

The use of a high-temperature, half-calcined dolomite cleanup process would result in a normalized efficiency of 0.85 at a fuel gas delivery temperature to the gas turbine of 1,610°F. About 4.5 percent of the coal heating value would be used to raise steam as the gas is cooled from 1,750°F, the gasifier exit temperature, to the best temperature for sulfur retention (1,610°F). Three low-temperature processes would use about 16.5 percent of the coal heating value to raise steam, if a fuel gas regenerator were used to raise the fuel delivery temperature to


AIR
INLET
LOW COMP.
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STEAM CYCLE
MAIN BOILER
SAT. STEAM
B.F.W.
STACK
M. P. BOILER
COAL
AFTER COOLER
BOOSTER COMP. (OPTIONAL)
COLD SIDE HEAT EXCH.
GASIFIER
H.P. BOILER
HOT SIDE FUEL REGEN.
CLEANUP
BOOSTER COMP. (OPTIONAL)
COLD SIDE FUEL REGEN.
STACK
L.P. PROCESS BOILER (OPT.)
HOT SIDE HEAT EXCH.
SECONDARY BURNER
FAN (MOTOR)
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Figure 5. Integrated COGAS/Coal Gasification Power Station

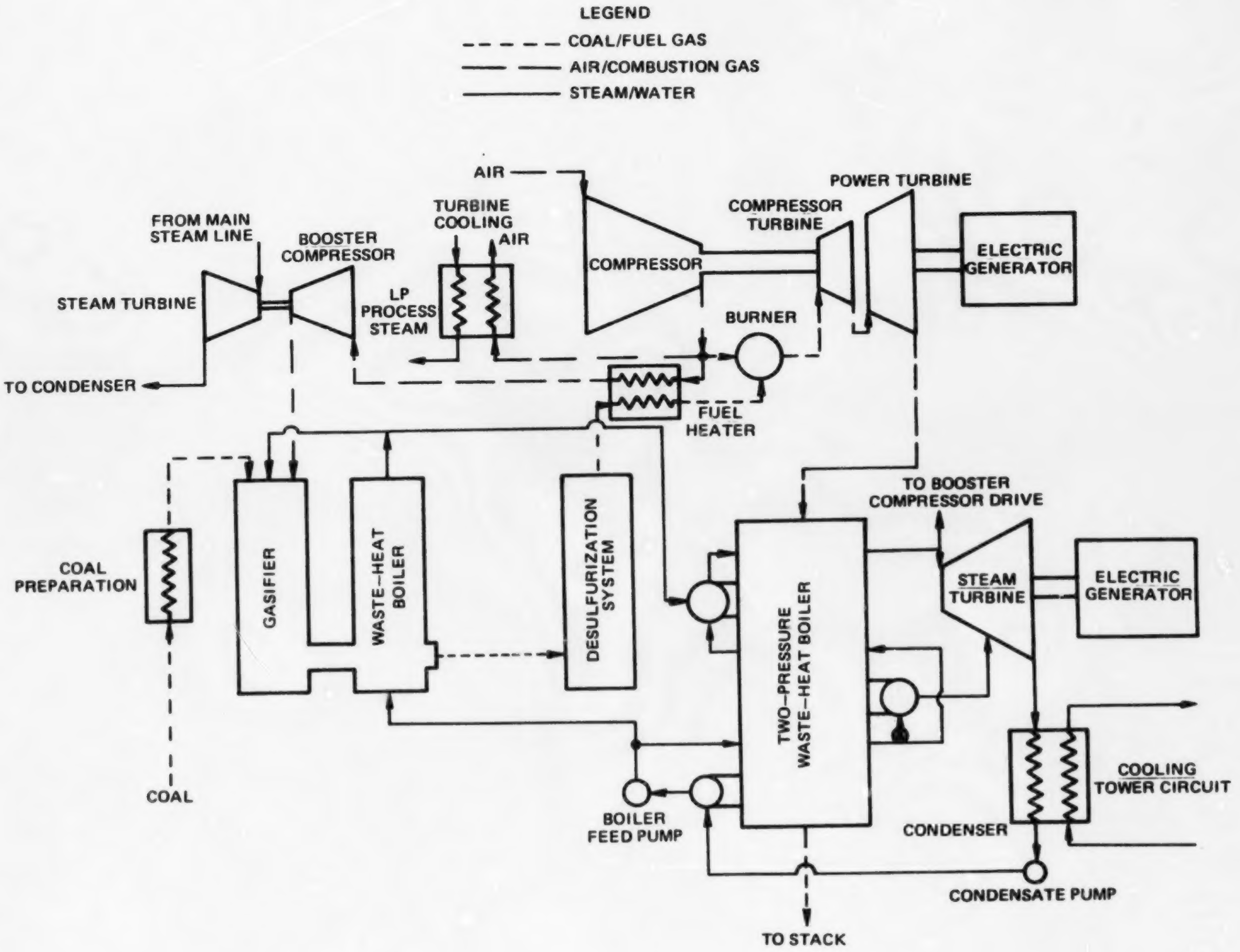
LEGEND
COAL/FUEL GAS
AIR/COMBUSTION GAS
STEAM/WATER
AIR
FROM MAIN STEAM LINE
BOOSTER COMPRESSOR
TURBINE COOLING AIR
STEAM TURBINE
LP PROCESS STEAM
COMPRESSOR
COMPRESSOR TURBINE
POWER TURBINE
ELECTRIC GENERATOR
BURNER
TO CONDENSER
FUEL HEATER
COAL PREPARATION
GASIFIER
WASTE-HEAT BOILER
DESULFURIZATION SYSTEM
TWO-PRESSURE WASTE-HEAT BOILER
TO BOOSTER COMPRESSOR DRIVE
STEAM TURBINE
ELECTRIC GENERATOR
COOLING TOWER CIRCUIT
CONDENSER
COAL
BOILER FEED PUMP
CONDENSATE PUMP
TO STACK

Figure 6. Integrated Coal Gasification—COGAS Power Station

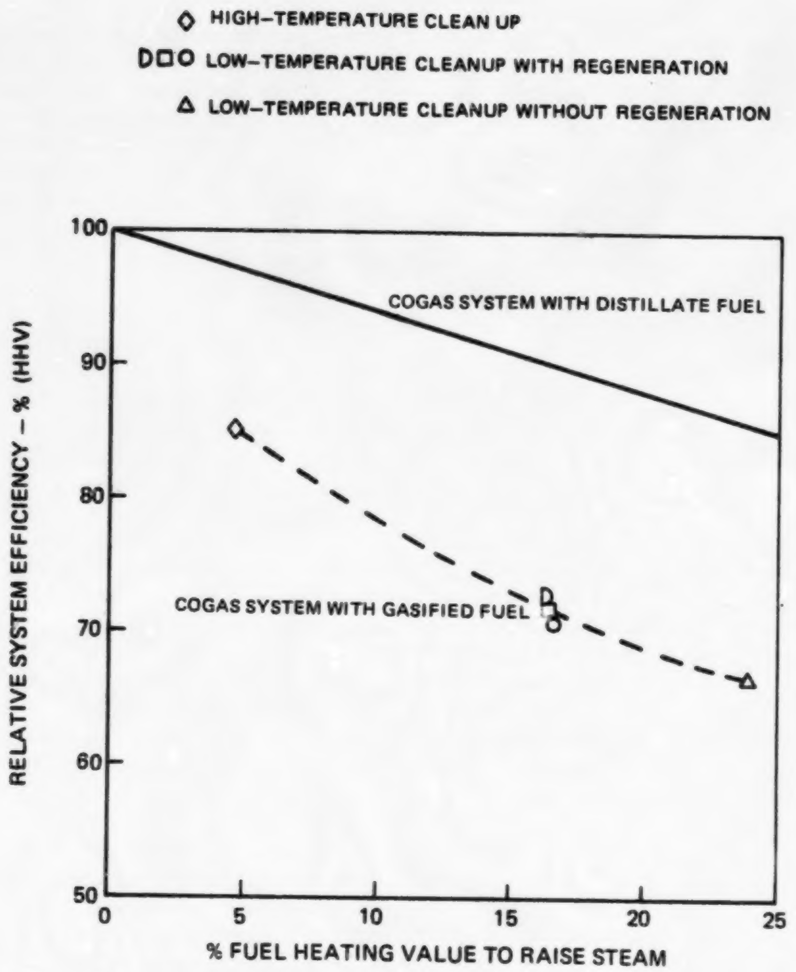

**Figure 7.** **Effect of Cleanup System Operating Characteristics On System Performance**

685°F. The normalized efficiencies of these systems are all about 0.72. If no regeneration were used, about 24 percent of the coal heating value would be used to raise steam, and the normalized efficiency would be about 0.67.

The differences in efficiency between the distillate-fired and gasified-coal-fired COGAS systems with high-temperature cleanup are due to gasifier heat losses (ash and cooling water), auxiliary loads for coal-handling equipment and the cleanup systems, and to the power required for the booster compressor. The COGAS systems with low-temperature cleanup face similar losses, plus degradation due to the large stream flow.

## ENVIRONMENTAL CONSIDERATIONS

The objective of all the foregoing has been to set the stage for discussions of the environmental impact of the gasified coal-fired combined cycle. At this time, the evaluations of this most important aspect of this advanced power cycle concept are just being initiated. The following comments, however, can be made on the major pollutant streams.

### *Sulfur Oxides*

As a practical limit, well within the EPA 1975 standards for coal-fired plants (1.2 lb $SO_2/10^6$ Btu input), a value of 100 ppm $H_2S$ + COS was set for the fuel gas leaving the cleanup system. This corresponds to less than 0.20 lb $SO_2/10^6$ Btu of coal, roughly a factor of six less than the standard. Table 5 shows the makeup of the fuel gas leaving three different types of low-temperature cleanup systems, while table 6 shows the fuel gas makeup from high-temperature systems. These tables also indicate the utility requirements for these systems.

If necessary, sulfur oxide emissions from the Claus plant also can be reduced to essentially negligible levels through the use of several commercially available processes (ref. 4).

### *Nitrogen Oxides*

The formation of nitrogen oxides is a much more complex problem than the formation of sulfur oxides. The nitrogen in the fuel gas in the form of $NH_3$, HCN, pyridenes, etc., contributes to $NO_x$ formation and should be removed in the fuel cleanup process. As has been commented previously, the low-temperature cleanup systems would remove the greater portion of these constituents, whereas it is not apparent that the high-temperature systems would do so (see tables 5 and 6).

The fuel delivery temperature also has a profound effect on the production of $NO_x$. In figure 8 (ref. 2) the combustion temperature, which determines the rate of $NO_x$ production, is plotted as a function of fuel gas heating value. Two curves are shown, one representing the increase in combustion temperature resulting from an increase in the chemical heating value of the fuel (the lower curve), and one showing the more rapid increase in combustion temperature with increased sensible heat (fuel temperature). If fuel gas having a 120 Btu/scf chemical heating value were sent directly to the engine from a low-temperature scrubber (e.g., 100°F as with an amine system), the adiabatic combustion temperature would be approximately 3,680°F. (The combustion air preheat due to compressor work also would effect the temperature). If a high-temperature cleanup system were used (1,750°F), the fuel chemical plus sensible heat would be increased to around 148 Btu/scf; and following the upper curve, a combustion temperature of about 4,250°F would result. Using the fuel gas regeneration scheme previously discussed, the final fuel temperature would be nearly 685°F, thus giving rise to a combustion temperature of approximately 3,900°F.

What effect on $NO_x$ production results from these varying combustion temperatures, and how do these rates compare with more conventional fuels? Figure 9 shows the $NO_x$ emissions as a function of combustion temperature for several turbine inlet temperatures. Assuming a 2,200°F (2,660 R) inlet temperature, the $NO_x$ emissions given in table 7 can be postulated.

The above projections are based upon a theoretical treatment developed by Pratt & Whitney Aircraft (ref. 5). These theoretical results have been borne out in tests on the combustion of low-Btu gas carried out by Pratt & Whitney Aircraft and the Turbo Power and Marine Systems, Inc. (subsidiary of United Aircraft) and Texaco, Inc. These tests (ref. 6) give $NO_x$ emissions from conventionally, designed, gas turbine combustors burning low-Btu fuel of the order of 5 to 10 ppm.

To the above values of $NO_x$ formed by atmospheric nitrogen must be added those values produced by the nitrogen-bearing compounds in the fuel. These compounds would be removed in the low-temperature cleanup systems (see table 5) but not in the high-temperature systems as presently configured. The amount of $NH_3$, HCN, HCNO, etc., formed is a function of nitrogen content of the various coals used. Using reference 2 as a guide for HCN, HCNO, etc., mole fractions of these constituents could be of the order of $10^{-5}$ to $10^{-4}$ or about 0.01 lb $NO/10^6$ Btu of coal. The $NH_3$ content shown in table 5 for low-temperature systems indicates concentrations varying from 0 to 600 ppm. This could add as much as 0.4 $lb/10^6$ Btu of coal for these cleanup systems. In table 6, $NH_3$ concentrations are shown to be six times larger, meaning that when both atmospheric and fuel nitrogen are considered, $NO_x$ emissions for high-temperature systems could be over 2 $lb/10^6$ Btu of coal, an unacceptable value. It is probable that not all the fuel nitrogen would be converted to $NO_x$, but at this time no reliable estimates of the fraction converted are available.

### *Trace Elements*

The fate of trace elements during gasification is difficult to assess. Work performed at IGT (ref. 7) provides the only source of data in the literature. Since these data are available, no further discussion will be attempted, except to note that the values of Pb, Ni, and V in the vapor state (probably as oxides) are of a magnitude that could potentially be harmful to the gas turbine (see table 2). These metallic vapors would condense during the low-temperature cleanup process and would not reach the turbine. It is not evident that the high-temperature processes would remove these potentially harmful fuel gas constituents.

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Table 5. Estimated performance of low-temperature cleanup of BCR gas

| Process | Polyethylene Glycol Ether | Hot Carbonate | Methanol |
|---|---|---|---|
| **Feed Streams**$^{(a)}$ | | | |
| BCR gas flow, mph | 426 | 426 | 426 |
| Temperature, F | 1750 | 1750 | 1750 |
| Pressure, psia | 450 | 450 | 450 |
| **Product Streams** | | | |
| Sulfur, lb/hr | 74.9 | 74.9 | 76.2 |
| Transport gas, mph | 34 | 34 | 34 |
| Product gas, mph | 333 | 350 | 330 |
| HHV, Btu/scf | 143 | 137 | 144 |
| Temperature, F | 100 | 250 | 90 |
| Composition, Vol % | | | |
| $N_2$ | 55 | 52 | 55 |
| CO | 19 | 18 | 19 |
| $CO_2$ | 9 | 7 | 8 |
| $H_2$ | 14 | 13 | 14 |
| $CH_4$ | 3 | 4 | 4 |
| $H_2O$ | 0 | 6 | 0 |
| $NH_3$ | | 0.06 | 0.06 |
| $H_2S$ + COS | 100 ppm | 100 ppm | 10 ppm |
| **Utilities** | | | |
| Cooling duty, MMBtu/hr | 2.9 | 3.3 | 3.0 |
| Steam @ 1300 psia, lb/hr | 107 | 107 | 107 |
| @ 65 psia, lb/hr | 1020 | 2505 | 780 |
| Electric Power, kw | 61 | 26 | 42 |
| Boiler Feed Water, lb/hr | 219 | 223 | 223 |
| Steam Condensate, lb/hr | 1233 | 2718 | 995 |
| Feed Gas Cooling, MMBtu/hr$^{(b)}$ | 4.9 | 5.2 | 4.6 |

(a) Based on 2000 lb/hr coal feed to gasifier

(b) Available for steam generation and/or gas reheat

## Table 6. Estimated performance of high-temperature cleanup and BCR gas

| | Sintered Iron Oxide | Molten Carbonate | Half-Calcined Dolomite | Fully-Calcined Dolomite |
|---|---|---|---|---|
| **Feed Streams**(a) | | | | |
| BCR gas flow, mph | 426 | 426 | 426 | 426 |
| Temperature, F | 1750 | 1750 | 1750 | 1750 |
| Pressure, psia | 450 | 450 | 450 | 450 |
| Dolomite, lb/hr | | | 33 | 273 |
| **Product Streams** | | | | |
| Sulfur, lb/hr | 69.3 | 71.8 | 67.2 | 74.8 |
| Transport gas, mph | 34 | 34 | 34 | 34 |
| Product gas, mph | 392 | 390 | 390 | 241 |
| HHV, Btu/scf | 125 | 125 | 125 | 143 |
| Temperature, F | 1000 | 1610 | 1610 | 1550 |
| Composition, Vol % | | | | |
| $N_2$ | 48 | 48 | 48 | 54 |
| $CO$ | 12 | 16 | 16 | 12 |
| $CO_2$ | 14 | 10 | 10 | 3 |
| $H_2$ | 17 | 13 | 13 | 3 |
| $CH_4$ | 3 | 3 | 3 | 21 |
| $H_2O$ | 6 | 10 | 10 | 6 |
| $NH_3$ | .38 | .38 | .38 | .44 |
| $H_2S + COS$ | .02 | .04 | .06 | .01 |
| **Utilities** | | | | |
| Cooling Duty, MMBtu | 1.9 | 1.2 | .5 | 1.0 |
| Steam @ 1300 psia | (1490.0) | 275 | (470) | (8103) |
| Electric Power | 98 | 25 | 22 | (322.0) |
| Boiler Feed Water | 3746 | 1113 | 606 | 8941 |
| Steam Condensate | 2183 | 130 | (122) | (430) |
| Process Water | | | 42 | 184 |
| Natural Gas | 595 | | | |

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(a) Based on 2000 lb/hr coal feed to gasifier

(b) Net product gas after deducting process fuel requirements

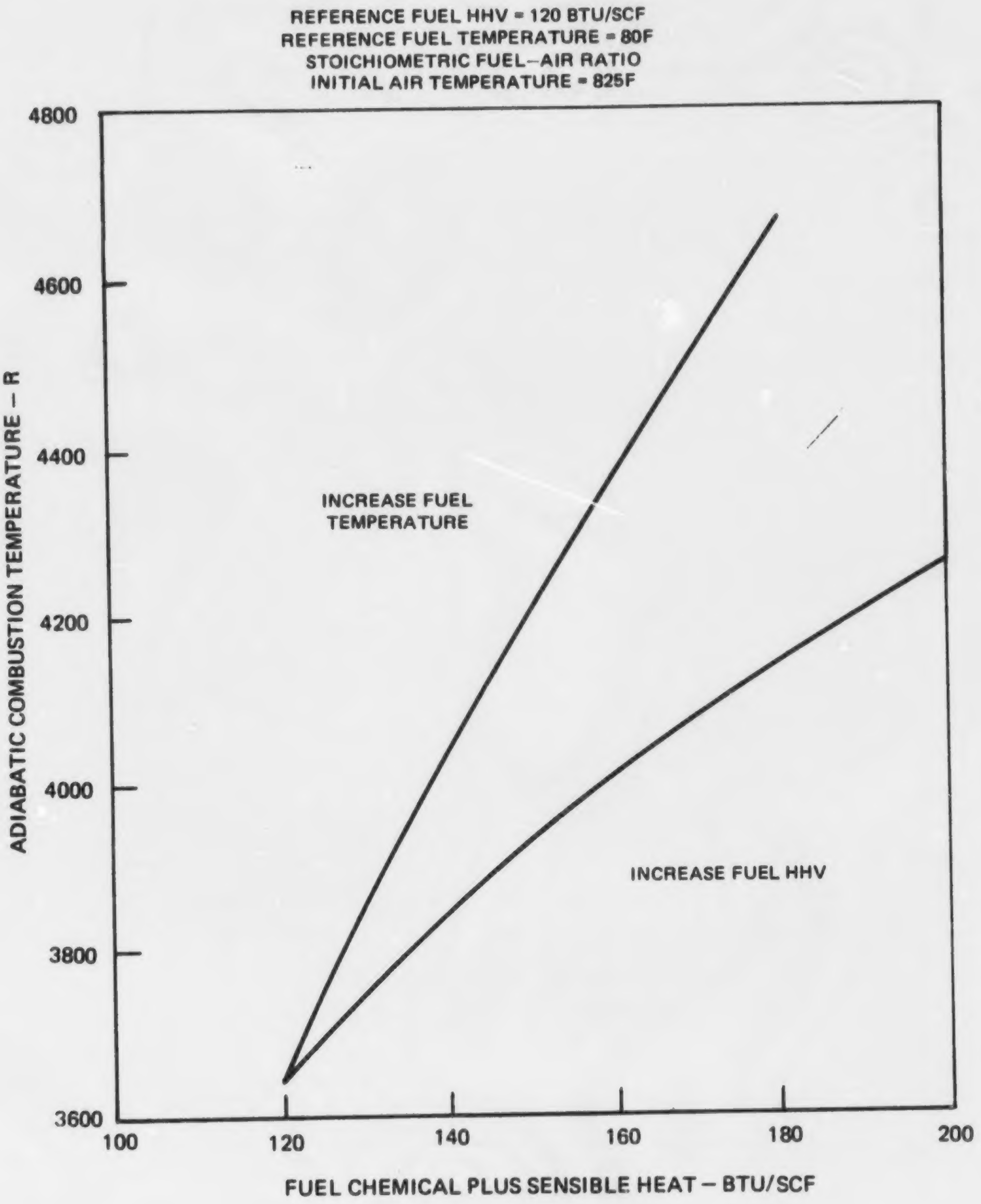

Figure 8. Effect of Fuel Gas Chemical And Sensible Heat On Combustion Temperature

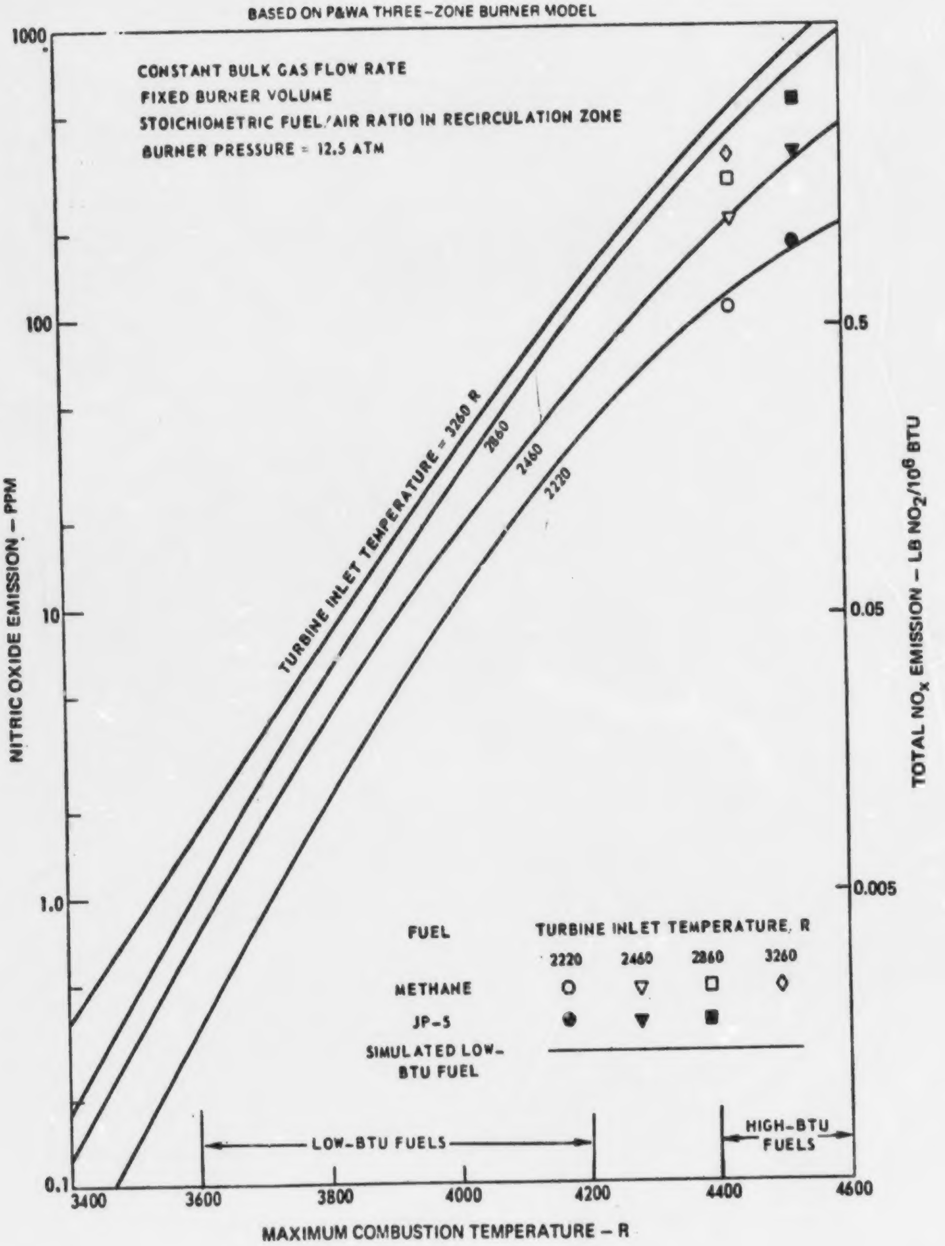

FIGURE 9. NITRIC OXIDE FORMATION IN GAS TURBINE BURNER

Table 7. Potential $NO_x$ emissions

| Type System | $NO_x$ Emissions[a] | | |
|---|---|---|---|
| | ppm | lb/$10^6$ Btu Coal[b] | lb/$10^6$ Btu Coal[c] |
| Low-temperature fuel | 2 | 0.01 | 0.02-0.4 |
| Low-temperature with regeneration | 8 | 0.03 | 0.03-0.4 |
| High-temperature fuel | 110 | 0.4 | 3.2 |
| Conventional distillate fuel | 450 | 1.0 | 1.0 |

(a) Based on gas turbine exhaust at approximately 300% theoretical air.

(b) $NO_x$ formed from atmospheric nitrogen.

(c) $NO_x$ formed from atmospheric and fuel nitrogen.

### *Thermal Discharge*

All of the power systems considered herein would be equipped with wet cooling towers; thus, there would be essentially no thermal pollution to surrounding water sources. The cooling tower systems would handle approximately 4,500 Btu/kWhr for the low-temperature systems and approximately 3,600 Btu/kWhr for the high-temperature systems. To these values must be added roughly 1,200 Btu/kWhr of heat rejected by the low-temperature cleanup systems and about 800 Btu/kWhr for the high-temperature systems.

## SUMMARY

The foregoing discussion has briefly described the results to date of a study being performed under EPA Contract 68-02-1099. The information presented should be treated as preliminary, since the analyses are continually being updated and the modeling techniques are being refined.

Table 8 serves as a method of summarizing the environmental impact of the integrated combined-cycle systems. There appears to be no difficulty in meeting the $SO_2$ limitations with the use of any of the low- or high-temperature cleanup systems. If a low-temperature system such as polyethylene glycol ether is used, essentially all the nitrogen-bearing compounds in the fuel gas would be removed and only $NO_x$ formed from atmospheric $N_2$ need be considered. Other low-temperature systems remove a portion of the fuel nitrogen and, even with regeneration, could still meet the environmental regulations. However, the high-temperature cleanup processes would not remove fuel nitrogen and appear to result in unacceptable emission levels. (A nitrogen-compound removal system could be added, but it would operate at 150 to 200°F, thereby negating any advantage of the high-temperature system.) Even when the emissions are considered on a lb/MWhr basis, which takes into account efficiency advantages, the high-temperature cleanup systems do not appear attractive from the environmental impact viewpoint.

Included in table 8 are two conventional steam stations to serve as a basis for comparison. The steam station using low-temperature cleanup would meet both the sulfur and $NO_x$ regulations, while the steam system using high-temperature cleanup would have difficulty meeting the $NO_x$ regulations.

Up to this point, no mention has been made of economics. Because of the status of the study, insufficient information has been obtained to generate reliable cost estimates. All that can be said at this time is that the COGAS systems continue to appear economically competitive with alternatiave clean power systems in the near term and, as technology improves, have the potential to become the lowest-cost (mills/kWhr) power generating system.

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Table 8. Summary of COGAS system environmental impact

| Type of Cleanup | lb $SO_2$ / $10^6$ Btu Coal | lb $SO_2$ / MwHr | lb $NO_x$ / $10^6$ Btu Coal | lb $NO_x$ / Mwhr | Btu to Cooling[(a)] / kwhr | Particulates |
|---|---|---|---|---|---|---|
| Low-Temperature | 0.15 | 1.7 | 0.02-0.4 | 0.2-4.0 | 4300 | None |
| Low-Temperature With Regeneration | 0.15 | 1.6 | 0.04-0.4 | 0.4-4.0 | 4500 | None |
| High-Temperature | 0.15[(b)] | 1.4 | 3.2 | 30 | 3600 | Environmentally Acceptable[(c)] |
| Conventional Steam with Gasification and Low-Temperature Cleanup | 0.15 | 1.8 | 0.01-0.2 | 0.1-2.0 | 7600 | None |
| Conventional Steam with Gasification and High-Temperature Cleanup | 0.15[(b)] | 1.7 | 3.2 | 32 | 7300 | Environmentally Acceptable |

(a) Only power system discharge is tabulated.
(b) Depending on process, this value could go to 0.6-0.8. Only power system emissions are tabulated.
(c) May not be acceptable to this turbine.

# ENVIRONMENTAL CONSIDERATIONS IN THE USE OF ALTERNATE CLEAN FUELS IN STATIONARY COMBUSTION PROCESSES

G. Blair Martin*

*Abstract*

*In response to the shortage of low-sulfur, premium quality fuels, such as natural gas and distillate oil, a variety of processes are under development for conversion of coal and other resources into gaseous and liquid fuels. The physical and chemical properties of these alternate clean fuels may be substantially different from the fossil fuels currently in use. The properties of these fuels can have an important impact on the pollutants emitted to the environment by combustion processes. The purpose of this paper is to review the available information on the combustion and emission characteristics of these fuels and to assess the applicability of combustion modification techniques for control of $NO_x$ and other pollutants. A comparison is made between chemical and physical properties of fossil fuels and selected alternate fuels, including low-Btu gas, methanol, and synthetic oil. Particular emphasis is placed on trace constituents of the fuel and the role of these constituents in pollutant formation. The similarities and differences of pollutant control techniques between alternate fuels and fossil fuels is considered. Finally, a summary of unresolved questions about use of alternate fuels in stationary combustion systems is posed.*

## INTRODUCTION

Within the past year, events have brought home the fact that clean fossil fuels are being used at rates exceeding our ability to produce them from existing sources. This realization has intensified efforts in the area of fuel conversion processes utilizing natural resources which are in abundant supply. One category of such processes is based on the conversion of coal to a variety of "clean" fuels. In this context, clean has come to mean low sulfur fuels. A variety of approaches are being taken, including the production of synthetic natural gas (SNG) which can be used to supplement natural gas in pipelines. In addition, various other conversion processes are being developed to produce other fuels such as low-Btu gas and synthetic oils. A second category is based on the conversion of oil shale to produce oil products. The fuel products of both categories of processes may have properties significantly different than those of current fossil fuels.

The fuels produced by these processes will be used either as substitutes for current fossil fuels in existing combustion processes or as the basis for design of new combustion processes to attain maximum efficiency. The different properties of these alternate fuels may have a significant impact on pollutant production in the combustion process and/or the energy efficiency of the process. The purpose of this paper is to assess available information on the properties of these alternate fuels and to discuss potential impact of these fuel properties on pollutant production and process efficiency of stationary combustion systems. The applicability of pollution control technology available and under development for conventional fuels may have an influence on the extent and/or necessity of fuel processing, thereby impacting both the cost and the energy efficiency of the fuel conversion processes. Primary consideration will be given to gaseous and liquid fuels for which the most information appears to be available. There are certain solid byproducts of the processes; however, the information on these is limited and will not be considered at this time. Several of the ideas are frankly speculative and are included to point out the possibility of basic rethinking combustion system design for alternate fuels, considering both environmental and efficiency aspects. This paper evaluates technical concepts and does not attempt to anticipate future EPA pollutant control regulations.

## BACKGROUND

To provide perspective for discussion of the environmental aspects of combustion of alternate fuels, it is necessary to provide some background on potential pollutant emission, available pollutant control techniques for conventional fuels, and comparison of properties of conventional and alternate fuels and classes of equipment in which these fuels may be used.

### *Identification of Pollutants*

In the combustion process, pollutants are produced

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during the oxidation of the fuel to release energy. Pollutants that are formed can be categorized into three general classes: (1) pollutants which are primarily related to the conditions of the combustion process; (2) pollutants which are primarily related to the composition of the fuels; and (3) pollutants which are attributable to a combination of the above.

*Pollutants related to combustion conditions.* Pollutants formed solely as a function of the conditions of the combustion process can generally be categorized as products of incomplete combustion, including carbon monoxide, unburned hydrocarbons, and carbon particulate. Formation of gaseous pollutants (carbon monoxide and unburned hydrocarbons) can be related to two areas of the combustion process: (1) air/fuel mixing at the burner, and (2) subsequent quench rate of the combustion products. If the proper burner design is used initially, complete oxidation of the hydrocarbon fuel will occur in the flame zone; however, the combustion gas may contain a substantial concentration of carbon monoxide. This carbon monoxide and any residual hydrocarbon will then oxidize in the high temperature postflame region unless the reactions are quenched. This quench can occur in the gas phase through the improper mixing of cold gas with the combustion products, or it can result from rapid cooling at the heat transfer surface. Therefore, control of these pollutants requires proper burner design which is also matched to the heat transfer configuration. Most properly designed fossil fuel systems produce CO and UHC levels below 100 ppm. However, after the system is placed in service, improper tuning or maintenance can have significant adverse effects on this performance. The other type of pollutant in this category is carbon particulate, which may form by either of two mechanisms. One of these is a gas phase particulate formation which tends to be a relatively small particle size formed by condensation of hydrocarbon radical fragments in the flame. This type of particulate would generally appear as "smoke" and can be formed for any fuel. The second type of carbon particulate is the larger size cenosphere particulate derived from coking of the fuel droplet or particle. This type of particulate is produced in combustion of liquid or solid fuels. In general, particulate formation must be prevented at the burner as it is extremely difficult to burn out in the postflame zone once formed.

*Pollutants related to fuel composition.* This category of pollutants is a function of fuel composition and is virtually independent of combustion conditions. The prime compounds which fall in this category are sulfur oxides, trace metals, and ash (noncarbon particulate). From all evidence, the levels of emissions of these compounds can be quantitatively calculated based on fuel composition and are not affected to any great extent by combustion zone conditions. (It is conceivable that the chemical form of trace metals could be altered; however, this has not been proven.)

*Combined effect pollutants.* Pollutants related to both combustion process conditions and fuel composition include nitrogen oxides, oxygenated hydrocarbons, and possibly carcinogenic polycyclic organic matter (POM). Of these three, the major information is available on the formation and control of nitrogen oxides. There are two potential paths for the formation of nitrogen oxide: (1) thermal NO, which is a function solely of the combustion conditions; and (2) fuel NO, which is a function of both fuel composition and local combustion conditions. Thermal NO is a product of high temperature fixation of atmospheric nitrogen present in the combustion air. This reaction set has an exponential dependence on temperature and a lesser dependence on oxygen concentration (ref. 1), although other factors enter in (ref. 2). Since practical turbulent diffusion flames are not characterized by a single unique temperature or oxygen concentration, the actual level of nitrogen oxide formed in any combustion process by atmospheric fixation is dependent on a variety of factors, including overall stoichiometric ratio, air/fuel mixing, and heat removal. These factors govern the location and size of local regions of high temperature in which thermal NO is formed. Thermal NO can potentially be formed in the combustion of any fuel, and it predominates for nonnitrogen-bearing fuels such as natural gas or distillate oil. Fuel NO is formed by oxidation of chemically bound nitrogen in the fuel to nitrogen oxides. The reactions forming fuel NO are apparently relatively independent of temperature and have a strong dependence on the availability of oxygen (refs. 3,4,5). The formation of fuel NO is also dependent on localized flame conditions. Fuel nitrogen is distinct from fuel sulfur in that it is not completely converted into nitrogen oxide, but rather, from the available evidence, some fraction is converted to nitrogen oxide with the balance going to molecular nitrogen. There has been unconfirmed speculation of formation of other nitrogen compounds such as hydrogen cyanide. The fraction of fuel nitrogen oxidized to NO is dependent on a number of factors, including overall excess air, level of nitrogen in the fuel, fuel injection method, and fuel/air mixing pattern. In general, the absolute level of fuel NO increases as the fuel nitrogen concentration increases. Evidence indicates that at least 50 percent of the total nitrogen oxide formed in the combustion of nitrogen-bearing fuels can be attributed to fuel NO. The fact that these two possible mechanisms for formation of NO exist requires the selection of the control techniques based on the nature of the fuel being considered.

The formation and control of the other two potential pollutants in this category, i.e., oxygenated hydrocarbons and polycyclic organic matter, is not well documented and must be considered in future research. The concern for oxygenated hydrocarbons arises primarily for fuels which have high levels of partially oxidized hydrocarbons (e.g., methanol) as fuel constituents. The consideration of POM in this category arises from the fact that many hydrocarbon constituents in heavier fuels are similar in structure to the multiple ring form of POM compounds. Also coal appears to produce high levels of POM. Both types of pollutants can also be synthesized in the combustion process by condensation of radical specie.

### *Combustion Control of Pollutant Formation*

Historically, combustion processes were operated to maximize combustion efficiency. This in turn meant minimum emissions of products of incomplete combustion, i.e., CO, unburned hydrocarbon, and carbon particulate. As a result, if modern combustion equipment is well maintained and operated, the level of emission of these pollutants is very low. For example, a utility boiler is normally operated at less than 200 ppm of CO in the stack.

Pollutants due only to fuel constituents are relatively insensitive to changes in the combustion process. For this reason, development of control technology for these pollutants, including $SO_2$ and metallic particulate, has been concentrated in two areas: (1) removal of these elements from the fuel prior to combustion, and (2) removal of the pollutants from the stack gases by scrubbing and/or precipitation. The major emphasis in fuel conversion processes currently under development is the removal of these elements prior to combustion. It is appropriate to mention here that through the years the Philosopher's Stone of combustion engineers has been a magic additive compound which, when added to the fuel in trace amounts, can remove sulfur oxides during the combustion process. To date, these pursuits have been relatively ineffective. The additives of this type which have been tried require several times stoichiometric amounts of material, provide limited removal of sulfur, and double or triple the ash loading in the flue gas.

In recent years, there has been a growing concern from the environmental standpoint about the emissions of nitrogen oxides into the atmosphere by combustion processes. Of the nitrogen oxides emitted into the atmosphere, approximately 50 percent originate from motor vehicle sources. The remaining 50 percent are generated predominantly from stationary combustion sources. Minor total contributions, which may be of considerable local importance, are emitted by nitric acid plants and other chemical processes. Development of combustion modification techniques for control of the formation of nitrogen oxide in the combustion process itself has been the main thrust of NO control research and development efforts in the United States. There are two mechanisms upon which modification techniques are based. The first mechanism is reduction of temperature in the combustion zone, thereby limiting the fixation of atmospheric nitrogen and the formation of thermal NO. This approach involves the introduction of an inert diluent into the combustion zone or the use of controlled rapid heat removal to accomplish the reduction of the temperature. The second mechanism is the limitation of local oxygen availability. This can have an effect on thermal NO; however, the predominant effect is on fuel NO. Although these two separate factors can be identified, essentially all practical combustion modification techniques embody both elements. A brief practical description of combustion modification techniques is given below.

The introduction of inert material into the flame zone can reduce the combustion temperature, thereby inhibiting formation of thermal NO. The two common techniques of introduction of inert are flue gas recirculation and water injection. Flue gas recirculation involves the removal of cooled flue gas from the stack of the combustion source at temperatures of 400° to 600°F* by a fan, and mixture of this flue gas with the incoming combustion air in proportion up to 30 percent. At 30 percent recirculation, the flame temperature is reduced by 600° to 700°F which, based on NO kinetics, should give significant reductions in NO. There is considerable experimental evidence that flue gas recirculation is an effective technique for control of thermal NO from clean fuels including natural gas and distillate oil (refs. 4,6). Reductions of NO up to 85 percent have been observed at 30 percent recirculation. The use of flue gas recirculation is considerably less effective for fuels which contain significant quantities of chemically bound nitrogen. Current evidence suggests that NO reductions of 30 percent can be obtained for heavy oil and lesser reductions for coal by the use of flue gas recirculation (refs. 5,7). This is attributable to the presence of fuel NO which is not significantly affected by reduction of flame temperature. Water injection can be accomplished by: (1) a direct spray of liquid water into the combustion zone, (2) the introduction of steam with either the fuel or the combustion air, or (3) emulsification of the water with liquid fuel. These techniques have been tried for both boilers and gas turbines (refs. 6,8,9). The results for both thermal and fuel NO are similar to those with flue gas recirculation.

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*NOTE: Although EPA's policy is to use metric quantitative descriptions certain nonmetric units are used in this paper for convenience. The following factors may be used to convert to the metric system:

1 Btu = 252.00 cal; 1 cu ft = 28.32 liters;
1 in. = 2.54 cm; 1 lb = 0.45 kg;
and 5/9(°F - 32) = °C.

Staged combustion is based on the operation of the burners in a fuel rich condition followed by delayed addition of secondary air to bring the overall stoichiometry to fuel lean conditions and complete the combustion. The effectiveness of this technique is based on a combination of factors. First, the peak temperature in the initial rich flame zone is reduced, and heat is removed prior to the addition of secondary air thereby reducing the peak flame temperature of the entire process relative to conventional combustion at the same overall stoichiometry. This provides a viable control technique for thermal NO. Second, the presence of the fuel rich flame zone limits oxygen availability and thereby provides an effective control technique for fuel NO. The major applications of this technique to date have been in utility boilers where significant reductions for all fuels have been achieved, particularly when using staged combustion in combination with low overall excess air (refs. 10,11). Staged combustion has not been applied to smaller sources due to process limitations making the additions of secondary air difficult. However, efforts are currently underway for the application of this technique to smaller boilers fueled by natural gas and residual oil (ref. 12).

Both of the techniques discussed above constitute what may be called external combustion modifications. A body of data is rapidly accumulating to indicate that burner design changes may in fact achieve the pollutant control objectives without the necessity for addition of either external inert injection or second stage air. Key variables in burner design are: (1) fuel injector type and configuration, and (2) method of introduction of combustion air. As with external combustion modification, the type of burner modification that is used depends on the nature of the fuel. For gaseous and clean liquid fuels, where the NO is predominately thermal, the design should promote relatively rapid air/fuel mixing and maximize entrainment of relatively cool combustion products from the recirculation zones near the furnace boundary heat transfer surfaces (ref. 13). For fuels containing significant quantities of chemically bound nitrogen, the burner design should promote local fuel rich zones in the flame to minimize the oxidation of the chemically bound nitrogen compounds and promote formation of $N_2$. The subsequent mixing of the bulk of combustion air with the fuel rich products to complete oxidation is controlled by burner design. For example, Heap has observed that if pulverized coal is rapidly mixed with combustion air by radial fuel injection, nitrogen oxide emissions of 800 parts per million are produced. Conversely, if the coal is maintained as a jet with limited primary air and surrounded by a flame front to delay mixing with the bulk of the combustion air, these emissions can be reduced to the level of 150 parts per million (ref. 14). It is obvious that burner design cannot consider merely nitrogen oxides. Other factors have to be considered including: stability of the flame, shape of the flame relative to the furnace volume, and combustion noise. All of these are criteria which must be considered for burner design.

Other control techniques which have been tried include reduced load, reduced air preheat, and other measures of this type. Although these measures are effective in reduction of NO (in some instances), they can also have an adverse impact on the overall process efficiency and/or output capacity and are therefore to be avoided whenever possible.

It is also recognized that in combustion processes the various problems are truly interrelated and, ideally, the use of processes which control nitrogen oxides should not result in emission of products of incomplete combustion being drastically increased and/or process efficiency being adversely affected. In general, the combustion modification techniques for control of nitrogen oxides are compatible with low emissions of products of incomplete combustion, if proper design is used. There is also evidence for staged combustion with utility boilers that the carbon efficiency, as based on residual carbon in the ash, is very slightly impacted, even when using a combination of staged combustion and low excess air to achieve control of nitrogen oxides from about 900 parts per million to 600 parts per million (ref. 11). Somewhat more difficult problems have been encountered with application of combustion modification techniques, particularly staged combustion to heavy oil in a smaller class of equipment; i.e., package boilers (ref. 15). In these experiments, an increase in smoke has accompanied significant decreases in nitrogen oxides. These problems should be overcome as a result of ongoing research.

The other consideration mentioned is overall process efficiency. Certain of the control techniques, particularly the ones involving inert injection and reduction of preheat, can adversely impact the overall process efficiency. A strong consideration in the EPA program for development of $NO_x$ control techniques has been to avoid any significant reduction in the process efficiency. This goal appears to be within the realm of the possible. Evidence on utility boiler testing suggests that the combination of staging and low excess air may, in fact, slightly improve the overall boiler efficiency (ref. 11).

### *Fuel Properties*

Based on the foregoing discussion, it is obvious that one of the first subjects that must be addressed when considering the environmental aspects of the combustion of fuels resulting from various fuel cleaning processes is the properties of those fuels themselves. Two areas of interest are: (1) trace constituents, such as nitrogen compounds, sulfur compounds, and metals present in the fuel product; and (2) any natural diluent in the fuel.

A summary of information for selected gaseous fuels is presented in table 1. The fuels represented are three classes of "synthetic" fuels: low-Btu gas, medium-Btu gas, and synthetic natural gas (SNG), with two types of natural gases shown for comparison (refs. 16,17). The only "trace element" information is on $H_2S$, which varies not only with process, but also with the parent coal. No information on nitrogen compounds or metals is available. There is some indication that a significant amount of ammonia is produced in some fuel conversion processes. It should be pointed out that these are compositions at the gasifier exit, without cleanup processes to remove $H_2S$ and other trace compounds.

A summary of information on selected liquid fuels is given in table 2. The "synthetic" liquid fuels include coal-derived oil, shale-derived oil, and methanol (refs. 16,18). The residual and distillate fuel oil properties presented for comparison are for the oils used in EPA/CRS in-house research and are "representative" of the fuel oils sold on the east coast. The coal-derived and shale-derived oils are uniformly lower in sulfur than the residual oil, but higher in nitrogen by factors of 3.5 to 7. The hydrotreated COED oil shows negligible levels of sulfur and nitrogen. The alcohol fuel properties are for pure methanol, as no information on "synthetic" fuels derived from coal was available. The alcohol fuels will almost certainly contain significant amounts of higher alcohols and possibly other hydrocarbons.

## EMISSION CHARACTERISTICS OF ALTERNATE FUELS

Information on the combustion and emission performance is unavailable for most of these alternate fuels. In many cases this can be attributed to the early state of development of the fuel conversion process. The emphasis to date has been on the properties of the fuel and the limited quantities of fuel which have been produced but have not been sufficient for combustion characterization. It is possible that some testing has been done and the results have not been published.

There is a scattering of information which is available on alternate fuels. In addition, there is a substantial body of information on combustion and emission characteristics of conventional fossil fuels. This section will attempt a synthesis of this information to give some indication of potential pollutant emission problem areas and to point out gaps in existing knowledge. The major discussion will center on the formation and control of nitrogen oxides since this is the area of combustion generated emission control about which the most is known. The experimental data shown in the figures has been recently generated, and more detailed presentations will be made at a later date. Other emissions are also considered, based on available information. In the following discussion, fuels having similar characteristics are considered as a group.

### *Synthetic Natural Gas*

The composition of SNG appears to be very similar to that of natural gas. The primary difference is the

Table 1. Comparison of various gaseous fuels

| Type of Fuel | Higher Heating Value Btu/FT$^3$ (Dry) | Gas Composition, Mole % | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | CO | $H_2$ | $CH_4$ | $C_2H_6$ | $CO_2$ | $N_2$ | $H_2O$ | $H_2S$ |
| Texarkana Natural Gas(a) | 967 | 0 | 0 | 96.0 | 0 | 0.8 | 3.2 | 0 | 0 |
| Cleveland Natural Gas(a) | 1131 | 0 | 0 | 80.5 | 18.2 | 0 | 1.3 | 0 | 0 |
| Synthetic Natural Gas Koppers-Totzek(b) | 962 | 0.08 | 2.5 | 94.2 | 0 | 0.4 | 2.8 | 0 | 0 |
| Synthetic Natural Gas Lurgi(b) | 980 | 0 | 0.8 | 96.8 | 0 | 1.2 | 1.1 | 0 | 0 |
| Medium Btu Gas Koppers-Totzek(b) | 298 | 50.4 | 33.1 | 0 | 0 | 5.6 | 0 | 9.6 | 0.3 |
| Medium Btu Gas Lurgi(b) | 302 | 9.2 | 20.1 | 4.7 | 0.5 | 14.7 | 0 | 50.2 | 0.6 |
| Low Btu Gas-Lurgi(b) | 180 | 13.9 | 19.6 | 5.5 | 0 | 13.3 | 37.5 | 10.1 | 0.6 |
| Low Btu Gas-Winkler(b) | 118 | 19.0 | 11.7 | 0.5 | 0 | 6.2 | 51.1 | 11.5 | 0.13 |
| Low Btu Gas-Hygas(b) | 236 | 13.5 | 16.6 | 8.4 | 0.6 | 12.7 | 28.9 | 18.2 | 0.8 |
| Low Btu Gas - U-Gas(b) | 150 | 17 | 11.6 | 4.1 | | 8.8 | 45.4 | 12.0 | 0.6 |

(a) Handbook of Chemical Engineering, John H. Perry-Editor, 3rd Edition.

(b) "Clean Fuels from Coal-Symposium Papers," Institute of Gas Technology, Sept. 10-14, 1973.

Table 2. Comparison of various liquid fuels

| Fuel | Higher Heating Value Btu/LB | Ultimate Analysis, Weight % | | | | | | API Gravity Degrees |
|---|---|---|---|---|---|---|---|---|
| | | C | H | N | S | O | Ash | |
| Distillate Oil[(a)] | 19,300 | 86.9 | 13.1 | 0.023 | 0.096 | 0 | < 0.002 | 35 |
| Residual Oil[(a)] | 18,980 | 86.8 | 12.5 | 0.22 | 0.89 | 0 | 0.03 | |
| COED Crude[(b)] | - | 83.0 | 8.4 | 1.1 | 0.35 | 7.15 | 0 | - 4 |
| COED[(b)] Hydrotreated | - | 88.0 | 11.6 | 0.01 | 0.01 | 0.38 | 0 | 24.5 |
| Synthoil[(b)] | 16,800 | 89.6 | 7.6 | 0.9 | 0.31 | 1.6 | (1.3) | 1.126 Sp.Gr. @ 60/60°F |
| H-Coal Crude[(b)] | - | - | 9.5 | 0.68 | 0.19 | - | - | 15 |
| H-Coal Low Sulfur Fuel[(b)] | - | | 8.4 | 1.05 | 0.43 | - | - | 4.4 |
| Toscoal Oil[(b)] | 16,200 | 80.7 | 9.1 | 0.7 | 0.2 | 9.4 | 0.2 | 4.5 |
| Shale Oil | 18,700 | 84.6 | 11.7 | 1.77 | 0.76 | | 0.01 | Sp.Gr. 60/60 0.903 |
| Alcohol Fuel | 9,578 | 37.5 | 12.5 | 0 | 0 | 50 | 0 | NA |

(a) CRS in-house fuels

(b) "Clean Fuels from Coal-Symposium Papers," Institute of Gas Technology, Sept. 10-14, 1973.

presence of 1 to 3 percent of hydrogen in the SNG. The only data on trace constituents show the hydrogen sulfide content of the SNG to be zero. There is no direct data on nitrogen compounds, although mention is made of ammonia as a byproduct stream from a scrubber used to remove $H_2S$ from the synthesis gas (ref. 16). This raises an interesting aspect of the environmental impact of the fuel conversion process as the possibility of using the ammonia as an onsite boiler fuel is mentioned. No details are given; however, it might be assumed that the ammonia would be fired in combination with a supplemental fuel, such as natural gas. Available small-scale combustion experiments indicate that, dependent on system design, substantial concentrations of nitrogen oxides could be present in the boiler flue gas (ref. 19). The conversion to nitrogen oxides would depend on ammonia concentration, identity of second fuel, point of $NH_3$ injection, overall excess air, and other factors; therefore, no quantitative assessment can be made. However, if all the coal nitrogen is converted to ammonia in the process and subsequently burned, the total NO emissions could equal or exceed those from direct combustion of the parent coal. The conversion can be limited by proper design of the combustion system, and this should receive careful consideration at any installation where disposal by combustion is anticipated.

The SNG should be indistinguishable from natural gas in conventional combustion equipment. The only exceptions may be slightly wider flammability limits due to the presence of hydrogen and the potential for fuel NO formation if any bound nitrogen compounds ($NH_3$, HCN, etc.) are present in the product gas. The principal source of NO would be thermal fixation, which can be controlled by any of the techniques previously mentioned. Products of incomplete combustion will not be significant with proper system design, as is the case with natural gas.

### *Medium-Btu Gas*

The synthesis gas fed to the SNG plant is generally a medium-Btu gas (about 300 Btu/ft$^3$) produced by an oxygen blown gasifier. The main constituents are carbon monoxide and hydrogen; however, large concentrations of water can also be present in the products of certain gasifiers. Although it is not normally considered in the United States, the possibility of firing this gas directly (after sulfur cleanup) does exist and should be considered here. The potential advantage compared to low-Btu gas is that smaller volumes of relatively nitrogen-free fuel gas need to be handled for the same Btu input to the furnace. The potential disadvantage is that the theoretical flame temperature is greater than that for methane, which could lead to high thermal NO emissions.

In this context a point worth considering is the use of this medium-Btu gas in a steam generator specifically designed to be oxygen fired. Since this could necessitate a considerable departure from current design practice, the final resolution will probably be based on economics as well as technical considerations. The advantages include smaller boiler size, higher combustion temperature, better heat transfer, and lower energy loss in the flue gas. Potential disadvantages are that problems may be encountered in finding economical materials and boiler construction to handle the higher temperatures, and the cost of the oxygen being used for combustion may be prohibitive. In the past, the cost of the oxygen appears to have been one of the main barriers to the concept; however, the presence of an oxygen plant on the gasification site and the projected higher cost of synthetic fuels may alter this conclusion. The impact of this concept on pollutant emissions is unknown at this time, but the following arguments can be considered. Even though the combustion temperature would be greater than 4,000°F, the thermal NO formation would be insignificant *if* the fuel gas were completely free of nitrogen. However, the analysis of the Koppers-Totzek gas shows about 1 percent $N_2$, and the possibility exists that this small amount of nitrogen could be converted nearly quantitatively to NO at these temperatures. Similarly, the nitrogen imputiries in the oxygen must also be taken into account. In addition the system would operate at initial temperatures at which the $CO/CO_2$ equilibrium is strongly shifted to CO. This would necessitate careful heat exhange design to avoid quenching too rapidly, resulting in large concentrations of CO, which represent both a pollutant and an energy loss from the system.

### *Low-Btu Gas*

The final gaseous fuel being considered is low-Btu which is produced in air-blown gasifiers. This fuel gas contains between 30 and 50 percent molecular nitrogen and has heating values between 100 and 240 Btu per cubic foot. The principal fuel constituents are CO, $H_2$, and $CH_4$, in proportions dependent on the specific process. Similar comprehensive information on trace elements other than $H_2S$ and COS is not available. The concentration of $H_2S$ must be reduced before the gas can be burned and, based on previous information on medium-Btu synthesis gas cleanup, it might be expected that nitrogen compounds and metals would be removed as well.

The other potentially significant variable is the fuel gas temperature as delivered to the burner of the combustion system using the low-Btu gas. This will depend on two factors: (1) the analysis of optimum gasifier/combustor system configuration based on an analysis of cost and energy efficiency; and (2) the development of high temperature (1,000°F) cleanup processes if these are warranted. (Note both of these aspects are being considered in ongoing EPA contracts with Ultrasystems and UARL, respectively.) The fuel gas temperature has a direct bearing on both furnace efficiency and pollutant emission.

Robson has reported that, if high temperature processes are used to remove the $H_2S$, the resultant clean fuel gas can contain 0.38 percent $NH_3$ (ref. 20). If this $NH_3$ were quantitatively converted to NO in the combustion process, the resultant fuel NO level would be 2000 ppm. The actual level of conversion would depend on the system design; however, for conventional practice a 50 percent conversion or 1000 ppm might not be unrealistic. The burner and/or system can certainly be designed to minimize the conversion; however, some further R & D addressed to this specific point may be required.

The theoretical flame temperature is a function of the inlet temperature of the fuel gas. If low-Btu gas temperatures of 70 and 1,000°F are assumed, the theoretical flame temperatures are approximately 3,000° and 3,450°F, respectively. As previously indicated, lower flame temperatures may result in lower NO emissions and thermal efficiency. For purposes of comparison the theoretical flame for natural gas is 3,550°F, and significant thermal NO formation can occur.

Another uncertainty with low-Btu gas is the effect on burner design, particularly fuel injector, and air/fuel mixing configuration. For methane combustion the fuel stream represents approximately 8 percent of the total air/fuel mixture, whereas, for low-Btu gas, the fuel gas is approximately 50 percent of the total. Also, for a constant injector area, the fuel gas velocity, which may be a significant factor influencing emission levels, will increase by a factor of approximately 3 if the gas temperature is raised from 70° to 1,000°F. No information is available on the effects of these burner parameters on combustion and emission performance of low-Btu gas.

The low-Btu gas produced specifically as a sulfur-free fuel derived from coal is envisioned as being burned onsite in two types of practical combustion systems, utility boilers and gas turbines. Although various forms of low-Btu gas produced as byproducts from industrial processes (e.g., blast furnace gas) have been burned for a long time in industrial boilers, either singly or in combination with natural gas, emission data for these practical combustion systems are not documented. It is fortunate that some experimental information has become available recently.

Under EPA Contract 68-02-0202, the International Flame Research Foundation of IJmuiden, Holland, has run a series of experimental trials to establish burner design criteria for control of $NO_x$ emissions from heavy oil and pulverized coal, with limited use of gaseous fuels for purposes of establishing baselines. In the AP-3 trial recently concluded, some runs with a simulated low-Btu gas were made (ref. 21). This gas was basically a blast furnace gas doped with approximately 6 percent methane to raise the heating value to about 180 Btu/ft$^3$, which is quite similar to the low-Btu gas composition produced by most gasifiers. This gas was fired singly and in combination with additional natural gas at a total heat input of 4.5 x $10^6$ Btu per hour with 15 percent excess air. The fuel injector and combustion air inlet areas were constant. The combustion air was introduced at 300°C and over a range of swirl values. The results for a highly cooled furnace are shown in figure 1. For these conditions, the NO emissions for 100 percent natural gas range from 47 to 62 ppm as a function of swirl; whereas, for 100 percent "low-Btu gas" the NO emissions were below the detection limit (4 ppm). These emission trends are consistent with the measured axial (maximum) temperatures of the combustion gases near the burner, which were 1,530° and 1,200°C, respectively. An interesting feature of the profiles is that the temperature difference decreases until at 2 meters it is only 100°C, suggesting that perhaps the heat release zone of the low-Btu gas is stretched out (i.e., the combustion intensity is reduced). Both decreased peak temperature and decreased combustion intensity would tend to produce low NO emission. It can also be seen from figure 1 that the first increment of low-Btu gas (33 percent of the heat input) has the largest effect on NO emissions (i.e., about 60 percent reduction), which is similar to results previously observed for external diluent addition by flue gas recirculation (ref. 6). CO emissions in the flue were comparable for natural gas and the low-Btu gas. Axial

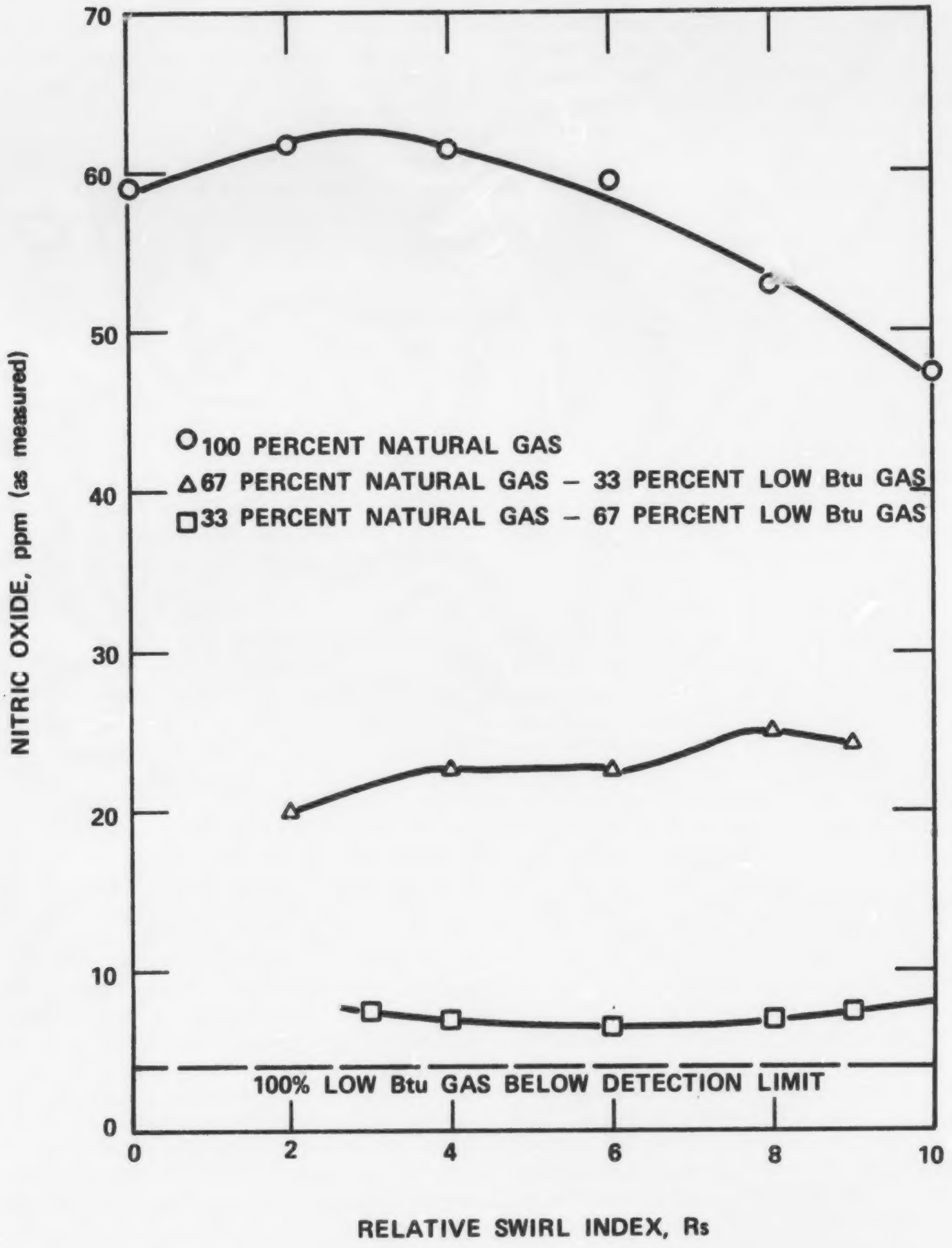

Figure 1. Nitrogen oxide emissions as functions of combustion air swirl and fuel gas composition

profiles of CO concentration were obtained and will provide further information on fuel burn-out and heat release patterns. The data is not available at this time.

The other application of low-Btu gas is to gas turbines for which a number of combustor inlet parameters are significantly different from those for boilers. A generalized summary is presented in table 3. Most of these conditions can have a significant effect on the emissions of NO. Fortunately, there has also been data recently published dealing with combustion of a low Btu gas in an experimental combustor simulating a combustor can (ref. 23). The combustor operated at low pressure (less than 40 inches of Hg absolute), but with air preheat up to 1,250°F to simulate compression heating. The observed NO levels for the low-Btu gas were zero to 10 ppm and for natural gas were 20 to 25. (It is not stated if the NO levels are as measured or corrected to some standard basis. It is noted that there was a 1.8 to 5.6 ppm contribution to these levels from the direct fired preheater.) Based on these numbers, Klapatch projects NO emissions from a TPM FTHC-1 (10-30 MW) gas turbine fired on low-Btu gas to be 50 percent and 65 percent less than the emissions for natural gas and No. 2 oil, respectively. The CO emissions for the experimental combustor on low-Btu gas were 3 to 4 times those for natural gas; however, it is also projected that proper design can produce equivalent levels for an actual engine. The stability limits for the low-Btu gas appear to be wider than those for methane.

Both sets of data tend to substantiate the projected low NO emission levels for combustion of low-Btu gas at 70°F.

### *Synthetic Oil*

The properties of the synthetic oil products in table 2 appear to be generally similar and, therefore, the discussion of coal-derived oils and shale oil will be covered as a single topic.

API gravity was the most commonly supplied physical property of the "crude" oils. The gravity was generally 5° API or less, which can be compared to a "typical" residual oil gravity of 12° API. This indicates a rather dense product which could be handled by a fuel system but would probably present flow difficulties. Viscosity information was presented in a wide variety of units and/or at several temperatures making comparison of flow properties laborious. The oil physical and chemical properties appear to vary as a function of parent coal and processing conditions. It may be assumed that at least limited processing will be necessary to obtain a suitable boiler fuel. One example of a hydrotreated oil produced a product upgraded to 24.5° API with essentially complete heteroatom removal (ref. 16). As with other aspects of the fuel conversion process, the amount of treatment given the "crude" oil will be dependent on the end use of the product, environmental considerations of use, and the economic factors involved in each of the two preceding factors.

The sulfur level in most of the crudes is less than 0.4 percent which is close to some current east coast sulfur regulations for residual oils (0.3 percent in some States). Economics might suggest combustion of these oils directly or with minimum treatment necessary to improve flow properties. From an environmental point of view, one of the problems then becomes fuel NO from the chemically bound nitrogen which ranges 0.7 to 1.77 percent. (This would correspond to 1,050 to 2,500 ppm NO if completely converted to NO and possibly 400 to 1,000 based on experience with actual levels of conversion.) The control of this fuel NO may be accomplished by staged combustion and/or burner design dependent on several factors. For convenience, chemically bound nitrogen can be divided into two

Table 3. Comparison of operating conditions for modern utility gas turbines and boilers firing natural gas

| Equipment Type | Boiler | Gas Turbines (22) |
|---|---|---|
| Pressure, atm | 1 | greater than 9 |
| Air preheat, °F | 600 | 600 or greater* |
| Overall excess air, percent | 10 | greater than 200 |

* Depends on Pressure Ratio and Use of Regeneration.

classes: (1) "volatile" compounds which are evolved early in the combustion process; and (2) "refractory" compounds which are burned out along with the char. These classes are intuitively defined, based more on speculation over experimental combustion results than by scientific tests. However, it should be mentioned that some analyses show that 50 to 60 percent of the nitrogen in high volatile bituminous coal is contained in the matter evolved during the ASTM standard "volatile" test (ref. 24). In the case of petroleum, the terms have somewhat less meaning as the nitrogen and sulfur heteroatoms tend to be quite refractory and concentrate in the heavy ends (e.g., residual oil) and any "volatile" nitrogen is determined by fuel pyrolysis in the flame. The available information does not give any indication of the distribution of nitrogen compounds in the synthetic crudes. This presents difficulties in assessing the potential effectiveness of control techniques.

In general, results of combustion experiments appear to fall into the two categories related to the nitrogen classes discussed above. In the first category are results of combustion of fuels with high levels of "volatile" nitrogen compounds. Much of the early work on control of fuel NO was based on distillate oil doped with relatively low boiling heterocyclic nitrogen compounds, such as pyridine (refs. 3,4). This represents a case of 100 percent volatile nitrogen, and conversion levels of 50 to 60 percent to NO were observed. When staged combustion techniques were applied, the level of fuel NO could be reduced from 750 ppm at 120 percent of theoretical air through the burner to about 100 ppm at 70 percent theoretical air through the burner and 120 percent overall (ref. 4). There is evidence to show that the predominant source of NO from coal is fuel nitrogen. Pershing has experimentally compared NO emissions from combustion of pulverized coal in air (1,030 ppm NO) to those for Argon/21 percent oxygen mixture (900 ppm) at 120 percent theoretical oxygen and has confirmed the hypothesis that fuel NO is relatively independent of flame temperature (ref. 7). Heap has shown that the NO emissions from pulverized coal can be reduced from 800 ppm to about 150 ppm by change in burner operating parameters and has proposed an hypothesis based on conversion of volatile fuel nitrogen compounds (ref. 14). In all of these cases of "volatile" fuel nitrogen, significant reductions in NO have been achieved by producing a fuel rich zone early in the combustion process by either "staged" combustion or alteration of the air/fuel mixing patterns by burner changes. These reductions in NO can apparently be achieved without apparent increases in carbon loss (smoke, CO, etc.). The results on the second category of fuels having refractory nitrogen (e.g., residual oil) are somewhat mixed. Turner has reported reductions of fuel NO of 40 to 50 percent (200 ppm fuel NO down to 100 ppm) in a refractory-lined package boiler without significant increases in smoke (ref. 5). On the other hand, Muzio has conducted experiments in a Dowtherm cooled unit (450°F wall temperature) and has encountered smoke problems, with staged combustion producing only 25-30 percent reduction in total NO. By combining flue gas recirculation and staged combustion, a 45 percent reduction in total NO was achievable (ref. 15). The results of Muzio may be a function of the burner configuration tested or of differences in the fuel used. In the AP-2 trials at the IFRF, Heap found the NO emissions for residual oil to be much less sensitive to burner parameters than coal was. The range of NO observed was from 250 ppm to 150 ppm at equivalent conditions (ref. 25).

These results may indicate the sensitivity of fuel NO emission from oil combustion to "volatile" nitrogen content (e.g., 85 percent reduction of fuel NO for doped distillate versus 25 to 40 percent for residual). In addition, they show that the production of carbon particulate (smoke) is dependent on system parameters and, possibly, on the fuel properties as well. As a result, all that can be definitely stated at this time is that combustion modification techniques are potential control methods for fuel NO from synthetic crudes from which significant amounts of fuel NO can be expected, and that the effectiveness will depend on the distribution of nitrogen compounds in the synthetic oils.

### *Alcohol Fuels*

Alcohol fuels have a number of uncertainties associated with them, including potential sources. The principal current source appears to be as an alternative to LNG; however, one or more fuel conversion processes producing alcohol fuels from coal, refuse, or other waste material seem to be a distinct possibility. The alcohol fuel may have a variety of compositions; however, a probable composition is 90 plus percent methanol with the balance composed of higher alcohols. From an energy standpoint, methanol ($CH_3OH$) can be viewed as a mixture of 47 mass percent distillate oil ($CH_2$) and 53 mass percent water. The energy content (HHV) per lb of $CH_2$ is then very close at 20,200 Btu for methanol versus 19,300 for a distillate oil. The other physical properties are obviously quite different with methanol boiling at 149°F versus an endpoint of over 700°F for a distillate oil. The "water" content of methanol is a disadvantage as it represents a mass of inert matter which must be transported and constitutes an additional source of heat loss in the combustion products. However, it may offer an advantage from the environmental aspects.

The author is conducting an EPA/CRS in-house experimental study on the characterization of combustion and emission characteristics of future fuels. The experimental system is a refractory wall combustor fired at a nominal 300,000 Btu per hour. Comparisons have been made between alcohol fuels and conventional fossil fuels. Some prelinimary data is shown in figure 2. The results shown are at 5 percent excess air without air preheat and with the refractory temperature above 2,500°F. Three features of the data are obvious: (1) the NO emissions for methanol are a factor of 4 less than distillate oil and a factor of 3 less than propane; (2) increasing the content of higher alcohols appears to increase NO emissions; and (3) although the alcohol fuels are liquid the emission trends with air swirl appear to follow those of gaseous propane.

For a distillate oil at 5 percent excess air, the nominal air to fuel mass ratio is 16. For the same heat input, methanol is equivalent to 16 masses of air, one of fuel, and one of water, or the inlet mixture is 5.5 percent "water." In this case, approximately a 66 percent NO reduction is achieved relative to propane. For comparison, Halstead achieved approximately a 65 percent reduction in NO by injecting 5 percent liquid water as a spray into a natural gas flame (ref. 6). The increase in percentage of higher alcohols also decreases the "diluent water" in the fuel, thereby increasing the theoretical flame temperature and increasing thermal NO. Finally, the appearance of the methanol flame was observed to be more similar to a gas flame (nearly nonluminous) than to an oil flame. This is probably due to very rapid vaporization of the methanol in the hot refractory chamber, coupled with lack of high boiling hydrocarbons which crack to form luminous soot particles. This behavior could account for the similarity of the trend of NO versus swirl to propane. Finally, since the "water" is not actually present in that form in the fuel, chemical effects may also play a role.

In all of these tests, the level of CO and unburned hydrocarbons was less than 30 ppm. However, it should be pointed out that under relatively cold wall conditions found in furnaces and boilers, the high "water" content of methanol could produce a quench effect leading to emission of products of incomplete combustion, particularly oxygenated hydrocarbons, unless the system is properly designed.

The potential energy inefficiencies associated with alcohol fuels are: (1) lower flame temperature limiting heat transfer; and (2) latent and sensible heat losses in the flue gas (e.g., for 20,000 Btu of methanol a 2 lb mass is required, 1 of which is "water." Considering only the 1,000 Btu/lb latent heat to form steam, at least 5 percent of the Btu is unrecoverable).

### *Combined Fuel Firing*

One concept has been to convert coal to a sulfur-free, low-Btu gas, which is burned in a boiler on the gasifier site. Dependent on the fuel conversion process and system optimization, the overall efficiency of the gasifier may result in an energy loss equivalent to at least 15 percent of the heating value of the coal (ref. 16), which is apparently prior to any fuel cleanup. The author would like to propose a concept which might satisfy environmental requirements and also offer an energy saving.

The current EPA New Source Performance Standard for sulfur oxide emissions from a utility boiler is 1.2 lb $SO_2$ per million Btu input. If a boiler fired with a combination of coal and low-Btu gas were to be regarded as a coal-fired power plant, the sulfur oxide standard could be met by firing 25 percent of the Btu input as a 3 percent sulfur coal with 12,500 Btu per lb or 50 percent of a 1 percent sulfur coal with 8,300 Btu/lb and the balance of the heat input as a zero percent sulfur low-Btu gas. The potential energy savings are 4 and 8 percent, respectively. This savings on energy input needs to be balanced against requirements imposed on particulate removal from the flue gas and other considerations.

Now let us consider the impact on other emissions. Although the data on the proposed fuel mix is not available at this time, some data has been generated in the recent AP-3 trial at the IFRF (ref. 21). Data on $NO_x$ emissions for 100 percent coal, 67 percent coal, and 33 percent simulated low-Btu gas, and 100 percent low-Btu gas were generated and are presented in figure 3. The low-Btu gas $NO_x$ emissions were less than 10 ppm at these conditions. If this is averaged on a 33 to 67 basis with the results of firing 100 percent coal, the result is the solid line (i.e., as if the fuels were fired separately). However, when the coal and low-Btu gas are mixed and introduced through the same injector, the lower curve results. Although this data is far from definitive and no attempt was

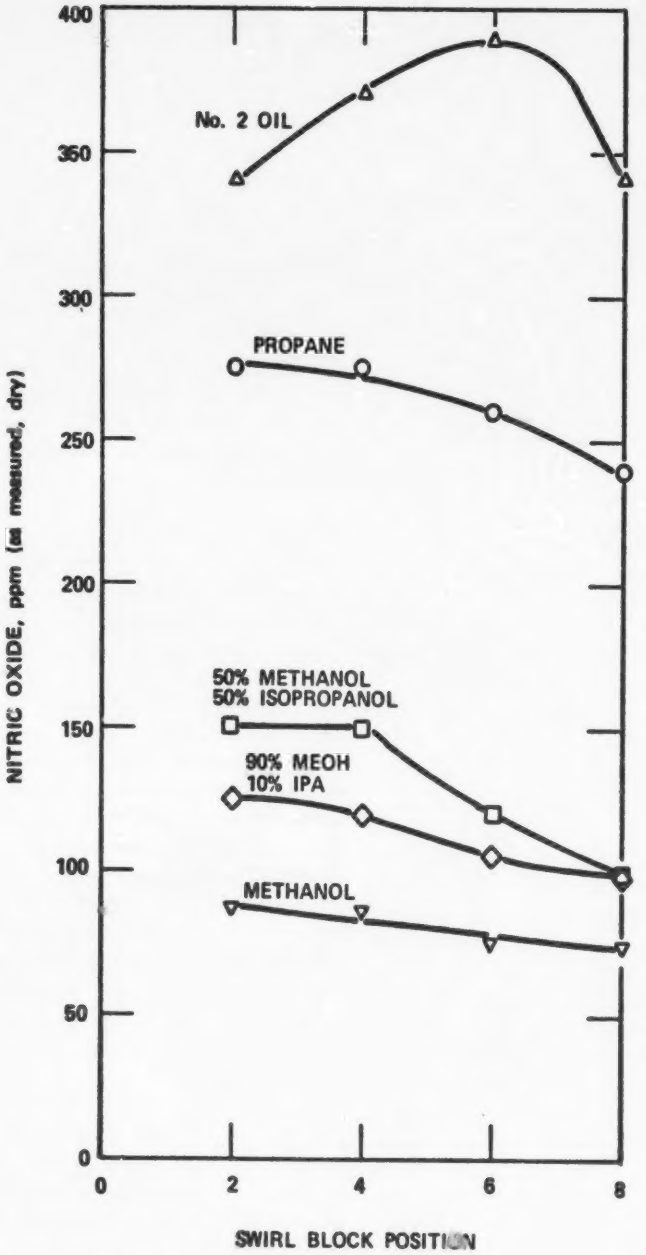

Figure 2. Comparison of nitric oxide emissions from combustion of distillate oil, propane, and three alcohol fuels

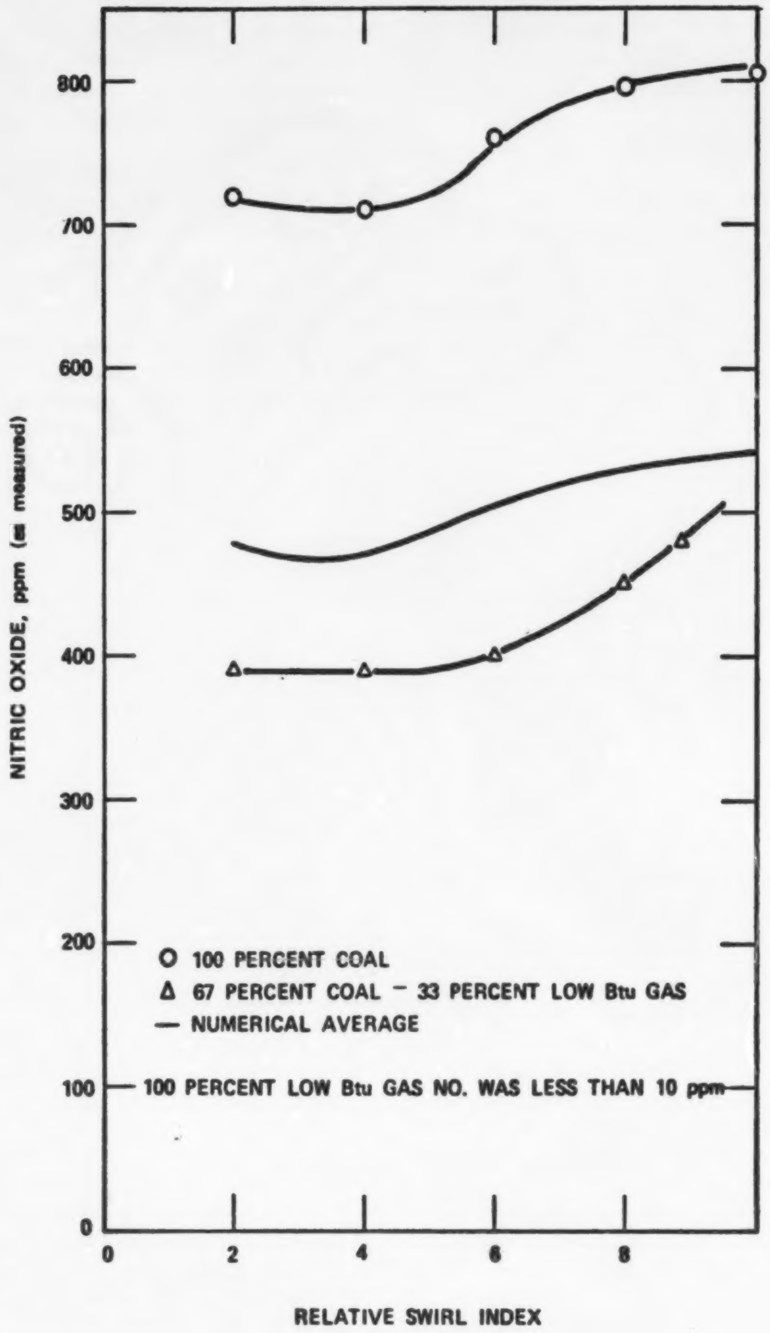

Figure 3. Nitrogen oxide emissions as a function of combustion air swirl and coal-blast furnace gas ratio

made to optimize the $NO_x$ reduction, it does indicate that combined firing of coal and low-Btu gas can actually reduce overall $NO_x$ emissions.

## SUMMARY

It is clear that the state of knowledge both of detailed composition of alternate fuels and of the environmental impact of utilizing these fuels in combustion processes is relatively limited. The main thrust of fuel conversion processes is to produce sulfur-free fuels for a variety of end uses and to remove ash as a part of the bargain. This reduces or eliminates two major pollutants normally produced in direct coal combustion. The pollutants which remain to be dealt with are nitrogen oxides, products of incomplete combustion, and, possibly, trace metals. Combustion modification appears to have good potential for control of nitrogen oxides for the alternate fuels which are not inherently low $NO_x$ emitters. Proper system design, based on combustion characteristics of the fuels, should eliminate products of incomplete combustion. Trace metals emissions will depend primarily on the characteristic of the fuel, and control will depend on removal at the fuel production site or from the flue gas.

To allow minimum environmental impact of combustion of alternate fuels, a body of information still remains to be generated, including the following:

1. Characterization of trace elements including nitrogen and sulfur compounds, and metals present in the full range of gaseous fuels.
2. For low-Btu gas, the effect of fuel temperature and air preheat on emission characteristics needs to be established. Information to allow burner and combustion system design for minimum pollutant formation compatible with maximum thermal efficiency must be established.
3. Quantitative information on the influence of high temperature $H_2S$ removal processes on the concentration of other trace elements in fuel gases needs to be established.
4. The fate of ammonia generated as a byproduct of fuel conversion processes and design criteria defined for low-NO combustion need to be established.
5. Characterization of the nitrogen and oxygen compounds contained in synthetic liquid fuels.
6. For synthetic crudes, combustion and emission characteristics as a function of fuel properties and extent of fuel processing need to be established. Special emphasis needs to be placed on evaluation of burner design and staged combustion for control of both NO and smoke.
7. The potential for formation of oxygenated hydrocarbons from fuel constituents and control requirements need to be established for all fuels, but especially alcohol fuels.
8. Further evaluation of combined firing of coal and low-Btu gas for pollutant control needs to be explored and system design criteria established.
9. Characterization and end use of combustible solid residue byproducts of the processes need to be established.
10. Critical analyses of the relationship of pollution control options to overall energy efficiency need to be carried out for each class of fuel.
11. The fact that unconventional fuels-generation processes are being developed, which may result in substantial increases in fuel cost, indicates that careful thought needs to be given to combustion system design with both environmental and energy efficiency considerations taken into account. On this basis, considering the systems as a whole, some dramatic departures from past practice may be justified.

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# STATUS OF FLUE GAS DESULFURIZATION TECHNOLOGY

**Frank T. Princiotta***

*Abstract*

*A comprehensive discussion describing the status of flue gas desulfurization (FGD) technology is presented. Information is included which lists operating and planned commercial FGD systems in the United States and Japan, $SO_2$ removal efficiencies achievable, typical capital and operating costs, other environmental considerations (e.g., sludge disposal), retrofit considerations, and barriers retarding rate of FGD system installation. Also included is a description of lime/limestone, double alkali, magnesium oxide, Wellman Lord and Catalytic Oxidation FGD systems including advantages, disadvantages, and operational status. Summaries of operational status of operating United States and selected Japanese FGD full-scale systems are also presented.*

## INTRODUCTION

Flue Gas Desulfurization (FGD) systems can be classified in two general categories: (a) systems in which the sulfur product is disposed of as waste (throwaway product systems), and (b) systems in which the sulfur product, such as sulfuric acid, is marketed (saleable product systems). The following commercially offered throwaway and saleable product systems are considered the most important for near term (through 1980) $SO_2$ control (ref. 1).

*Throwaway Product Systems*
- Lime Scrubbing
- Limestone Scrubbing
- Double Alkali

*Saleable Product Systems*
- Magnesium Oxide Scrubbing
- Wellman Lord (Sodium Sulfite Scrubbing)
- Catalytic Oxidation

An FGD system which best meets the specific requirements of a given $SO_2$ control problem can be selected from this list of available systems. (A description of each of these systems is presented in appendix A. The information in appendix A includes process descriptions, history and operating experience of the systems, advantages and disadvantages of the systems, and $SO_2$ removal efficiencies. Also, a performance and reliability summary is included for each system.)

## OPERATING AND PLANNED COMMERCIAL FGD INSTALLATIONS IN THE UNITED STATES

Along with the advance of FGD technology over the last 5 years has come substantial utilization of FGD systems in the United States and Japan. In the United States, FGD systems are already operating on 2,000 MW of the 18,000 MW of electric generating capacity for which FGD is planned. Figure 1 shows operating and planned power plant capacity controlled by FGD systems. Table 1 summarizes ordering trends for the various FGD control systems. These systems are planned for plants using high-sulfur content Eastern coal, low-sulfur content Western coal, and high-sulfur oil. (Appendix B briefly describes all known operating and planned full-scale FGD installations for power plant $SO_2$ control in the United States.) Based on the ordering trends suggested by these data, the following conclusions were made:

1. Although orders for FGD systems to date have been significant, the installation rate is still demand-limited;
2. 18,000 MW of generating capacity represent only a small fraction of the capacity which can and should be controlled by retrofitting FGD systems to existing plants; there is a potential demand for the installation of FGD systems on approximately 50,000 MW of generating capacity in 1975;
3. Based on the number and variety of FGD systems which have started up and the additional systems that will commence operation within the next 12 months, a substantial operational data base is being generated.

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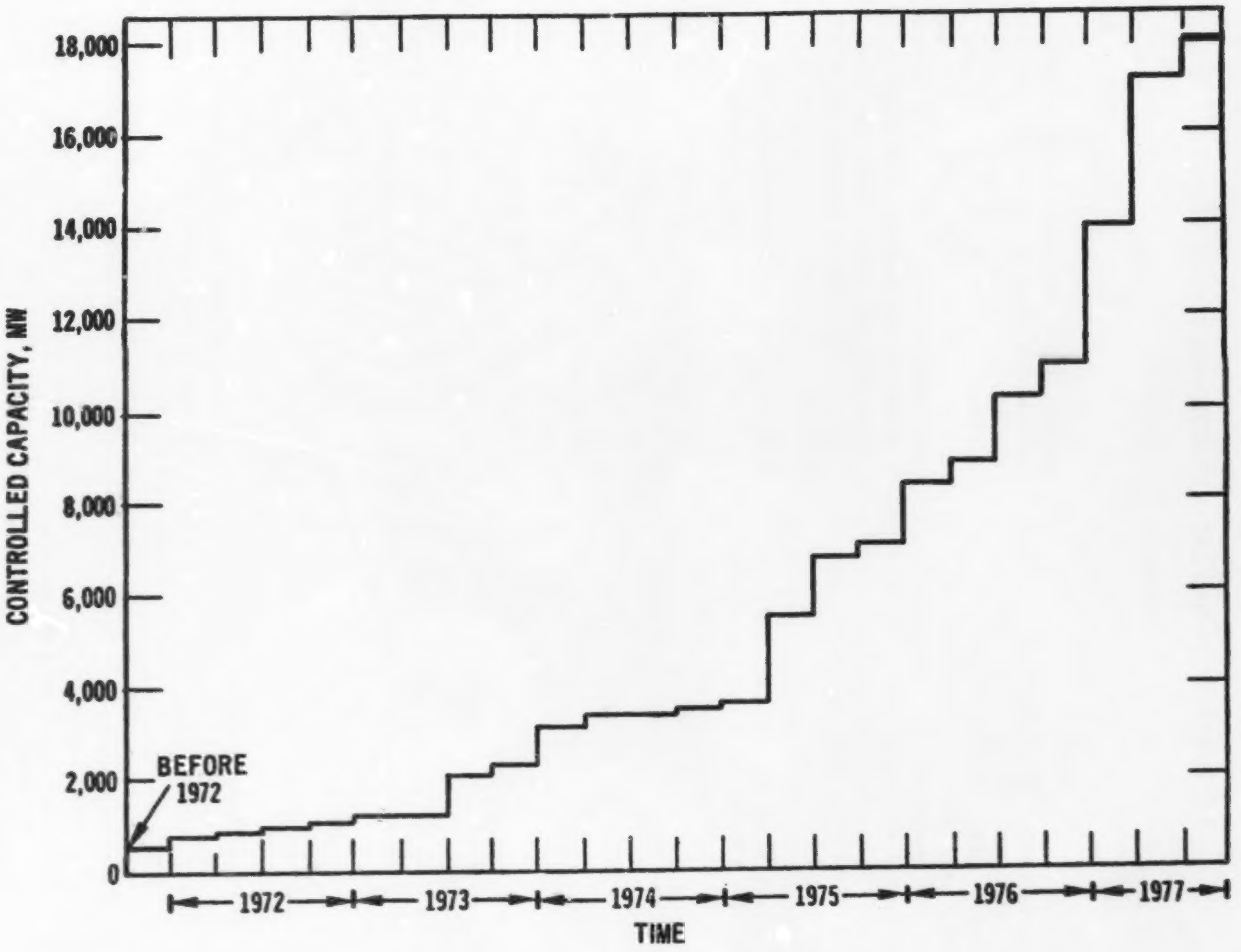

FIGURE 1. CUMULATIVE U.S. POWER PLANT CAPACITY WITH INSTALLED $SO_2$ CONTROL

Table 1. Summary of Operating and Planned Flue-Gas-Desulfurization Systems on U.S. Power Plants as of September 1973

| FGD System Type | No. of Units | Total Mw | Started Up No. | Started Up Mw |
|---|---|---|---|---|
| Limestone (LS) | 9 | 3931 | 3 | 1076 |
| Lime (L) | 11 | 3945 | 4 | 725 |
| L/LS - Not Selected | 10 | 5929 | | |
| Magnesium Oxide | 3 | 370 | 2 | 250 |
| Other $SO_2$ Control Systems | 5 | 600 | 1 | 110 |
| Process Not Selected | 5 | 2960 | | |
| Totals | 43 | 17735 | 10 | 2161 |

## OPERATING AND PLANNED COMMERCIAL FGD INSTALLATIONS IN JAPAN

As a result of increasingly more stringent $SO_2$ regulations in Japan, the rate of installation of FGD systems has accelerated during recent years. In Japan, utilization of these systems is considered a viable and cost-effective means of achieving standards set by regulations for the predominantly oil-burning utility industry. Presently, more than 60 commercial and prototype FGD plants are in operation. Even though most of the plants are of relatively small capacity and are designed to treat waste gas from industrial boilers, chemical plants, and smelting plants, several large FGD systems have been installed. Table 2 summarizes ordering trends for selected $SO_2$ control systems for Japanese boilers. (Appendix C briefly describes these units.) Presently, several of these installations are generating important performance and operability information. As is the case for U.S. installations, a wide variety of process types have been selected, and much of the information being obtained from them is relevant and important with respect to the application of FGD in the United States.

## FGD RELIABILITY SUMMARY

The reliability of currently available systems has been the subject of some question since $SO_2$ control systems must exhibit the high degree of reliability required by the utility industry. The required reliability has been achieved in Japan and will be achieved in the United States with the early resolution of a number of applications and engineering problems related to specific hardware components and system design parameters. Solutions to each of these problems have been developed and demonstrated at one FGD installation or another. It should be noted that the above assessment of technology status is consistent with that stated by the Sulfur Oxide Control Technology Assessment Panel (SOCTAP) which consisted of representatives from the Council on Environmental Quality, the Office of Science and Technology, the Department of Commerce, the Federal Power Commission, and the Environmental Protection Agency (ref. 2). Since the Panel's findings were reported in late 1972, further operating experience at Boston Edison, Mitsui Aluminum and Japan Synthetic Rubber units,

Table 2. Summary of Selected $SO_2$ Control Systems on Japanese Boilers (As of September 1973)

| Process | No. of Units | Total Mw (Planned & Operating) | Operating | |
|---|---|---|---|---|
| | | | No. | Mw |
| Wellman-Lord | 4 | 545 | 2 | 295 |
| Dilute Sulfuric Acid-Gypsum (Chiyoda) | 3 | 536 | 1 | 52 |
| Double Alkali-Gypsum (Limestone) | 3 | 456 | 1 | 156 |
| Mitsubishi-JECCO Lime/Limestone-Gypsum | 4 | 530 | | |
| Wet Limestone | 1 | 100 | | |
| Carbon Adsorption | 1 | 139 | 1 | 139 |
| DAP-Manganese | 1 | 110 | 1 | 110 |
| Lime Scrubbing | 1 | 156 | 1 | 156 |
| | 18 | 2572 | 7 | 908 |

Louisville Gas & Electric system, and Arizona Public Service tend to corroborate these findings. The state of the art of $SO_2$ desulfurization technology has advanced rapidly over the last year and many full-scale installations have been ordered and some operated in the United States and Japan. (A summary of operating experience to date for most of the full-scale units which have started up in the United States and Japan is presented in appendix D.)

The operating experience at two plants with throwaway products and two plants with saleable products is considered particularly significant. The most successful operation of a throwaway system has been the Chemico calcium hydroxide scrubber process in Japan. It has operated on a coal-fired boiler at the Mitsui aluminum plant in Japan since March 29, 1972, without any significant downtime; availability of this unit has been effectively 100 percent since start-up. Sulfur dioxide and particulate removal efficiencies have been approximately 85 percent and 98 percent, respectively. In addition, there are important similarities between this application and typical U.S. requirements, including:

1. Retrofit on an existing coal-fired boiler;
2. Installation on a moderately large boiler (156 MW);
3. Availability of calcium hydroxide in the United States.

The system takes on additional significance since it was based on U.S. technology (Chemico) and a similar unit, installed at Duquesne's Phillips Station, is presently in a start-up phase.

A second significant throwaway product system is the Louisville Gas & Electric FGD system, which started up in April 1973 on a 70 MW generating unit at the Paddy's Run Station. Since start-up, scrubber availability has been high with no evidence of major operating problems. This is considered further evidence that FGD technology is a workable and viable $SO_2$ control technique.

In addition to the two full-scale throwaway FGD facilities discussed above, EPA's prototype (three 10 MW units) lime/limestone scrubbing facility at the TVA Shawnee Steam Plant has also provided valuable reliability information. Limestone reliability verification testing has identified scrubber types and operating modes which have allowed reliable operation for up to a month's duration; long-term limestone reliability testing has recently been initiated. Also, testing at Shawnee and at a supporting EPA pilot facility at EPA-Research Triangle Park (RTP) in North Carolina has generated important process chemistry information which indicates that enhanced reliability and operability of lime and limestone systems can be achieved under selected operating conditions.

Of the saleable product systems, the Wellman-Lord regenerable sodium sulfite scrubbing process has operated most reliably to date. A system treating flue gas at Japan Synthetic Rubber's Chiba Plant has shown reliable and efficient operation for almost 3 years (since June 1971), producing high quality sulfuric acid. The main disadvantage of this system is the requirement for discarding a sodium sulfate bleed stream which is ecologically and economically undesirable. However, there are indications that bleed rates can be substantially decreased so that less than 5 percent of the sulfur in the incoming flue gas need be discarded, compared to the present 10 percent. Also, a larger 220 MW Wellman-Lord system has been operating reliably in Japan at a Chubu Electric utility boiler since May 1973 under varying load (peaking) operation. No significant reliability problems have been encountered, and availability has been close to 100 percent.

Chemico's magnesium oxide, saleable product system at Boston Edison's Mystic Station was started up in April 1972 and operated until June 1973 on an intermittent basis due to mechanical difficulties. However, during June and July 1973, operability was greatly improved and availability was greater than 80 percent until a scheduled boiler outage terminated the run. The system was recently restarted and, after some minor mechanical problems, has operated at an availability greater than 80 percent since February 22, 1974. Sulfur dioxide removal efficiencies have been in excess of 90 percent with no significant scrubber problems. Recent experience with the critical regeneration system has been good. There appears to be a high probability for long-term, reliable operation of this unit in the near future. It should be noted that no significant environmental problems have been identified with this system. However, problems in marketing large quantities of sulfuric acid will probably limit acceptability of saleable product systems to only a portion of the total potential flue gas desulfurization market.

## $SO_2$ REMOVAL EFFICIENCIES

When evaluating $SO_2$ removal efficiencies, it should be noted that a removal efficiency of about 75 percent is needed to meet the New Source Performance Standards while burning bituminous coal containing 3 percent sulfur. Generally, efficiencies of 85 percent are sufficient to meet the $SO_2$ emission limitations of most State implementation plans.

As discussed above, a number of FGD systems are being tested and evaluated. At the Mitsui aluminum plant near Omuta, Japan, the Chemico scrubbing unit has exhibited reliable, essentially trouble-free operation, with removal efficiencies of 80 to 90 percent since March 29, 1972 (ref. 3). The Wellman-Lord scrubbing unit, at the Japan Synthetic Rubber plant near Chiba, has accumulated over 15,000 hours of operation since June 1971, with a removal efficiency averaging about 90 percent.

In the United States, $SO_2$ removal efficiencies of approximately 90 percent have been reported for Boston Edison's magnesium oxide and Louisville Gas & Electric's calcium hydroxide FGD systems. Commonwealth Edison has reported efficiencies of 80 to 90 percent, although this limestone scrubbing facility has been plagued with mechanical difficulties since its start-up in February 1972.

Based on the removal efficiencies reported at various facilities, it is apparent that such systems are capable of meeting all present $SO_2$ emission regulations.

## COSTS

It is difficult to generalize on control costs since costs must be calculated for a specific application and usually cannot be readily extrapolated to predict costs for other applications. This is the case because of varying interrelationships between the many factors which influence costs. For control processes, these factors, all of which affect capital and operating costs, include: the size of the power plant (whether the system is new or retrofit), amounts of sulfur and ash in the fuel, pollution control requirements, price of reactants, solid waste disposal constraints, and scrubber type.

Table 3 presents ranges of capital and annualized costs for the important FGD processes. For comparison purposes, capital and operating costs for a coal-fired power plant and a range of low-sulfur cost increments are included. As can be seen, FGD costs are not particularly sensitive to FGD process type. Generally, the aforementioned application factors have a more significant effect. In general, incremental capital costs for including a FGD system in the construction of a new electric generating plant range from a low of $30 to a high of $50/kW capacity. This includes particulate control equipment, where required. The average incremental cost for new generating plants is expected to be about $40/kW.

Capital costs for retrofit installations to existing generating plants in most cases are expected to be in the $45 to $65/kW range. For some retrofitted plants, installation costs have been estimated as high as $80/kW or more. However, the practical limiting cost for retrofitting is fixed by economic considerations at each particular plant.

The total annual costs estimated for stack gas cleaning range from 1.5 to 3.0 mills/kWh, (ref. 4) with a mean of about 2.0 mills/kWh; this is comparable to the cost increment associated with low-sulfur fuels. The as-produced cost of electricity is about 9 mills/kWh, whereas the price to customers averages about 20 mills/kWh. It is estimated that the average increase in electricity costs to consumers will be about 3 to 6 percent assuming 100,000 and 200,000 MW of installed FGD capacity by 1983, respectively. However, for those utility systems which are predominantly coal users with essentially the total capacity controlled by FGD, price increases could be as high as 15 percent.

Since FGD annualized costs are comparable to the low-sulfur fuel cost increment and are a reasonable fraction of electrical generating costs, FGD costs, although significant, are not considered prohibitive.

## OTHER ENVIRONMENTAL CONSIDERATIONS

One of the major considerations inherent in any FGD system is the necessity to dispose of or utilize large quantities of a sulfur-containing product which may be throwaway or saleable. To date, most attention has been focused on lime/limestone scrubbing systems which produce throwaway sludges. The disposal of these sludges adds to the existing problem of flyash disposal for coal-fired power plants.

Two environmental problems are associated with throwaway sludges: (a) potential water pollution problems primarily associated with the dissolved species in the liquid phase of the sludge; and (b) potential land utilization problems since nonsettling sludges make land reclamation difficult. However, techniques are available which should minimize adverse environmental effects. These techniques include: (a) operating the scrubber in a close-loop mode (returning all liquid streams back to the scrubber circuit to reduce the water pollution potential); (b) using pond liners in closed-loop systems employing well-engineered disposal sites to eliminate water pollution problems; and (c) employing commercially available sludge fixation processes to convert the sludge into a more desirable landfill material with acceptable structural properties and decreased permeability and leachability.

## RETROFIT CONSIDERATIONS

Recent surveys have estimated the proportion of existing boiler capacity having sufficient space between the boilerhouse and stack to allow installation of $SO_2$ scrubbing equipment. Generally, a scrubber, reheater, induced draft fan, and ducting must be installed in this limited area. Approximately 20 to 25 square feet of ground space per megawatt is required for installation of the various $SO_2$ control systems currently commercially available. Boilers built within the last 10 years tend to be relatively large units. Approximately 85 percent of this boiler capacity less than 20 years old or greater than 100 MW can be retrofitted. Older and smaller boilers are less likely to have available space and are generally peak loaded. Fortunately, their size and low load factor account for a small proportion of total $SO_2$ emissions.

Process equipment outside of the scrubber area (hold tanks, pumps, etc.) is of less concern to the retrofit problem since it can be located in the peripheral areas of the plant. However, disposal of throwaway sulfur products can be troublesome, especially in metropolitan areas where land costs may be prohibitive. Generally, about 0.27 acre-ft/yr of ponding is required for each megawatt of boiler capacity. Several techniques have been proposed for disposing of large quantities of throwaway sulfur products. To date, most of the operating lime or limestone scrubbing systems have relied on disposal of the sludge materials in a disposal pond on the power plant site. If sufficient land is available, the pond is designed to eventually store all of the solid

## TABLE 3. COMPARISONS OF $SO_2$ CONTROL PROCESS SYSTEMS FOR COAL-FIRED POWER PLANTS

| | Reactant input requirements | Throwaway or recovery | Approx. invest. costs[a] for coal-fired boilers $/kw | Approx. (annual) costs,[b] mills/kw-hr | | $SO_2$ removal efficiency,% |
|---|---|---|---|---|---|---|
| | | | | No credit for S recovery | With credit for S recovery | |
| Coal-fired power plant | N. A. | N. A. | 200 | 8.9[c] | N. A. | N. A. |
| Low-sulfur fuel increment (coal and oil) | N. A. | N. A. | N. A. | 2.0 - 4.0 | N. A. | N. A. |
| Wet lime/limestone/ $Ca(OH)_2$ slurry scrubbing | Lime (100-120% Stoich.); limestone (120 -150% Stoich.) | Throwaway $CaSO_3$/ $CaSO_4$ | 35 - 52 | 1.5 - 2.4 | N. A. | 80 - 90 |
| Magnesium oxide scrubbing | MgO alkali; carbon and fuel for regeneration and drying | Recovery of conc. $H_2SO_4$ or elem. sulfur | 36 - 66 | 1.6 - 3.0 | 1.4 - 2.8 | 90 |
| Monsanto catalytic oxidation (add-on) | Catalyst $V_2O_5$ (periodic replacement) and fuel for heat | Recovery of dilute $H_2SO_4$ | 43 - 67 | 1.6 - 2.7 | 1.5 - 2.6 | 85 - 90 |
| Wellman-Lord process (soluble sodium scrubbing with regeneration) | Sodium make-up and heat for regeneration | Recovery of conc. $H_2SO_4$ or sulfur | 40 - 68 | 1.5 - 3.2 | 1.2 - 2.8 | 90 |
| Double alkali process | Sodium make-up plus lime/limestone (100-130% Stoich.) | Throwaway $CaSO_3$/ $CaSO_4$ | 26 - 47 | 1.2 - 2.2 | N. A. | 90 |

(a) Generally, where a cost range is indicated, the lower end refers to a new unit (1000 Mw); the high end refers to a 200 Mw retrofit unit. Costs include particulate removal and are in 1973 dollars.

(b) Assumptions: Costs calculated at 80% load factor, fixed charges per year ~18% of capital costs.

(c) Includes environmental controls to minimize land and water pollution.

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waste produced during the power plant lifetime. Such ponds are fed by a bleed stream which is pumped either directly to the pond or via a thickening system (clarifier, filter, centrifuge) with the thickened sludge pumped to the pond. The supernatant liquor from both the dewatering system and the pond is usually returned to the scrubber circuit.

Another important disposal technique, used where land is not available at the plant site, involves maximum dewatering of the throwaway bleed stream using one of the many effective combinations of clarifying, filtering, or centrifuging equipment available; the solid, dewatered sludge can then be transported to a suitable landfill site. However, some sludge materials are difficult to dewater mechanically. Although sludge products which retain large quantities of liquor are difficult to transport and eventually lead to land-use problems due to the instability (nonsettling) of the wet sludges, chemical fixation processes are commercially offered by several firms including Dravo Corporation and IU Conversion Systems, Inc.

## BARRIERS TO SUPPLYING, INSTALLING, AND OPERATING FGD SYSTEMS

There are a number of institutional barriers in the electric utility and control systems industries which affect the rate of application of $SO_2$ control systems. Some of the most important are: (a) the inadequacy of the market demand to encourage development of a supply industry; (b) the limitations in vendor and construction capabilities when (and if) the market situation becomes supply limited; (c) the necessity to maintain adequate electrical reserve generation margin; (d) the lack of process chemistry expertise in the electric utility industry; and (e) fuel switching alternatives where higher costs for low-sulfur fuels can be passed directly to consumers by means of fuel adjustment clauses. The cost of control systems cannot be directly passed on in a similar manner. These factors tend to delay the ordering, fabricating, assembling, and placing into operation of $SO_2$ scrubbing systems.

An important factor now restricting system installation is the currently limited market demand for $SO_2$ control systems. This lack of demand by the electric utilities and other industries arises from such factors as lack of confidence in the ability of the systems to perform as promised, anticipation that regulations may be altered in the near future, potential difficulties in raising capital and obtaining rate increases to cover expenses for pollution abatement, and the lack of suitably trained personnel in the industry to evaluate and operate these systems. With increased demand pressure, scrubber systems could be constructed at a significantly higher rate than at present.

When and if the market situation becomes supply limited, the following are among the factors which will limit the rate of FGD system installation: qualified vendors and their capacities, availability of construction workers and critical components (fans, pumps, etc.), production capacity, capital availability, and limitations imposed by reserve margin considerations. Based on qualified vendor capacity, which was considered the major limiting factor, the SOCTAP (ref. 2) estimated that FGD systems could be installed on 10,000 to 20,000 MW of generating capacity by 1975 and that 48,000 to 80,000 MW could be controlled by 1977. Based on delays in ordering equipment to date, actual numbers will probably be closer to the low end of these estimates.

Nationally in the electric power industry, there is an upper limit to the generating capacity which can be retrofitted each year because of the necessity to maintain adequate reserve margins. The upper limit is related to the fact that it takes up to 6 weeks to tie in a scrubber to an existing boiler, necessitating downtime for that period; this factor may preclude higher, more desirable rates of installation. In particular, there may be scheduling problems in retrofitting scrubbers in the middle central and middle southern parts of the country where the large coal-fired utilities, already under pressure because of delays in installing new equipment, are concentrated. Electric utility companies must also plan for the loss of 2 to 6 percent of plant power output with use of a control system.

The electrical utility industry has little expertise in large-scale chemical process technology. Thus, there may be serious operational problems once the scrubbers are installed because of the lack of familiarity with the operational details of the scrubbing system. The utilities now depend almost completely on control system vendors and engineering consultants for technical advice.

There is a major economic deterrent to the installation of stack gas scrubbers. The utilities can meet the $SO_2$ standards by converting coal-fired plants to low-sulfur oil, or by burning low-sulfur coal. Both options have broad implications for national economic and environmental policies. Many utilities are allowed to pass most of the increased costs for low-sulfur fuels directly and immediately on to the consumer without regulatory commission action. On the other hand, utilities must apply for rate increases to cover the capital and operating expenses of the scrubbers. It would appear reasonable and desirable to pass FGD system costs directly to the customer as are the increased costs for low-sulfur fuels.

## CONCLUSIONS

FGD is the only significant, near-term alternative to the use of scarce, low-sulfur fossil fuels in many power plants. The costs for the installation of FGD systems are not considered prohibitive; annualized costs are comparable to the cost increments associated with the use of low-sulfur fuel for many applications. Since FGD technology is applicable for the control of $SO_2$ emissions from the vast majority of existing and new power plants, it is considered an important factor in helping achieve the country's clean air goals consistent with the requirements of the Clean Air Amendments of 1970.

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## APPENDIX A

## DESCRIPTION AND SUMMARY OF IMPORTANT FGD SYSTEMS

### A. THROWAWAY PRODUCT SYSTEMS

### 1. *LIME AND LIMESTONE FLUE GAS DESULFURIZATION PROCESSES*

*Process*

Several methods have been developed for the use of limestone and lime slurries in a wet scrubbing process. The major variations are as follows (see fig. A-1):

1. Use of limestone ($CaCO_3$) added to the scrubber circuit;
2. Use of hydrated lime ($Ca(OH)_2$) added to the scrubber circuit;
3. Use of limestone injected in the boiler effecting calcination to lime with subsequent lime slurry scrubbing.

In all three process modes, a slip stream consisting of reaction products, flyash, and unreacted alkali is subjected to a dewatering operation and is discharged as waste to either a disposal pond or landfill site.

The overall reactions for limestone and lime scrubbing can be represented by the following reactions:

*Limestone:* $CaCO_3 + SO_2 + \frac{1}{2}H_2O \rightarrow CaSO_3 \cdot \frac{1}{2}H_2O + CO_2$ (1)

*Lime:* $Ca(OH)_2 + SO_2 \rightarrow CaSO_3 \cdot \frac{1}{2}H_2O + \frac{1}{2}H_2O$ (2)

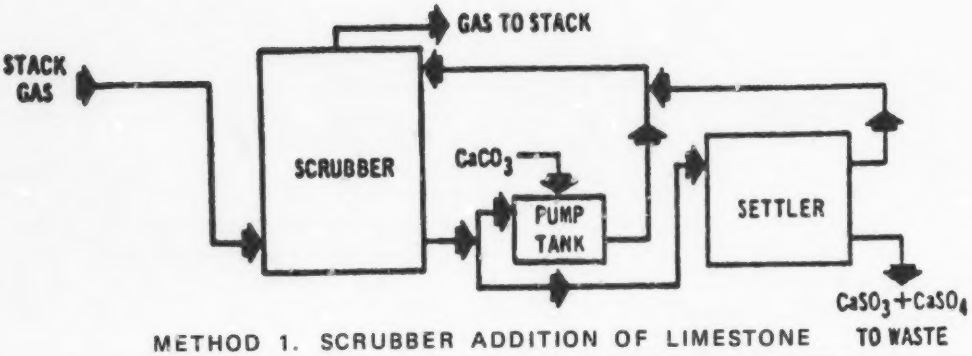
*History and Experience*

There has been a considerable amount of bench model, pilot plant, and full-scale activity in FGD since the early 1930's. Over the last 5 years, developmental activities have been particularly intense, and 31 full-scale commercial systems have been ordered since 1968 in the United States of which 7 have started up and have a backlog of operating experience.

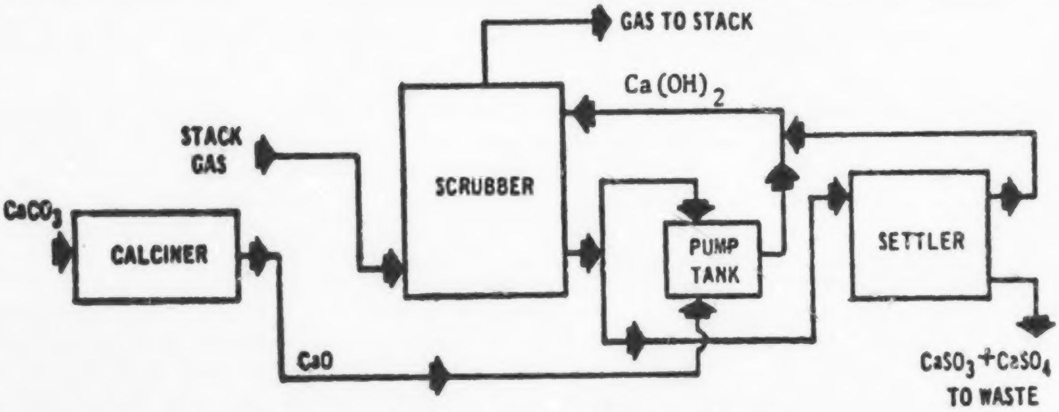
The boiler injection plus the wet scrubbing process, has been extensively tested on a commercial scale by Combustion Engineering, Inc., (C.E.) since 1968. After 4 years of intermittent operation due to numerous technical difficulties, C.E. no longer offers this process.

The first full-scale installation in this country to introduce limestone into the scrubbing circuit was the 175 MW Commonwealth Edison Will County Station, Unit No. 1. This unit was started up in February 1972, and has operated intermittently since then. It has achieved $SO_2$ removal efficiencies in the range of 75 to 85 percent. The major problems experienced to date are demister pluggage with a soft, mud-like substance, lack of system reliability due to mechanical problems, and waste sludge disposal. A great deal has been learned in the past year at the Will County Unit) concerning practical operating problems with limestone scrubbing. None of the problems encountered at this unit appear to be insurmountable.

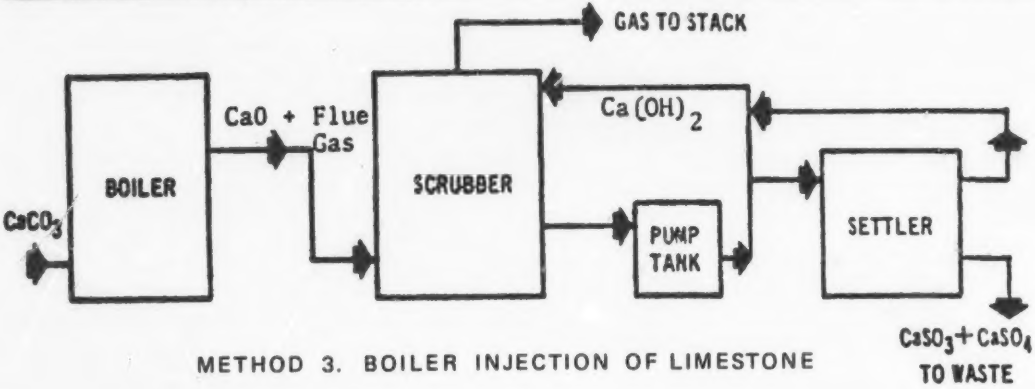
Additional valuable information concerning operation of lime/limestone scrubbing processes is being gathered at the versatile EPA prototype test facility at TVA's Shawnee Steam Plant. The 30 MW facility includes three types of 10 MW (equivalent) scrubbers (venturi, TCA, and marble bed), extensive process instrumentation, and sophisticated data acquisition and handling systems. Tests to date have


GAS TO STACK
STACK GAS
SCRUBBER
$CaCO_3$
PUMP TANK
SETTLER
$CaSO_3+CaSO_4$ TO WASTE

METHOD 1. SCRUBBER ADDITION OF LIMESTONE


GAS TO STACK
$Ca(OH)_2$
STACK GAS
$CaCO_3$
CALCINER
SCRUBBER
PUMP TANK
SETTLER
CaO
$CaSO_3+CaSO_4$ TO WASTE

METHOD 2. SCRUBBER ADDITION OF LIME


GAS TO STACK
BOILER
CaO + Flue Gas
SCRUBBER
$Ca(OH)_2$
$CaCO_3$
PUMP TANK
SETTLER
$CaSO_3+CaSO_4$ TO WASTE

METHOD 3. BOILER INJECTION OF LIMESTONE

FIGURE A-1. MAJOR PROCESS VARIATIONS FOR USE OF LIME OR LIMESTONE FOR REMOVAL OF $SO_2$ FROM STACK GASES.

identified test conditions which have led to reliable operation during tests of up to 1 month in duration.

The most recent successful operation of a lime wet scrubbing process is the 70 MW installation at Louisville Gas & Electric Company's Paddy's Run Station. The unit uses carbide sludge ($Ca(OH)_2$) as the alkaline absorbent. No scaling or plugging problems have been encountered in over 1,000 hours of closed-loop operation since April 1973. The system has demonstrated near 100 percent availability while removing 85 to 95 percent of the $SO_2$ in the flue gas from boilers fired with coal containing 3.5 to 4.0 percent sulfur. Waste sludge is thickened, filtered, and disposed of as untreated landfill.

The most successful lime scrubbing system is Chemico's Mitsui Aluminum Company's facility. This 156 MW power plnat has been retrofitted with two Chemico dual-stage venturi scrubbing systems, each capable of handling 75 percent of the full-load gas flow. The system has demonstrated reliable, trouble-free operation since being put on stream in March 1972. The plant is presently burning 2 percent sulfur coal (1,800 to 2,200 ppm inlet $SO_2$) and achieving 80 to 85 percent $SO_2$ removal from the flue gas using carbide sludge as the alkaline absorbent. Since coming on stream, the system has operated at near 100 percent availability. The absence of scaling difficulties has been attributed to operational know-how developed by Mitsui in extensive pilot plant tests (in Japan) and precise pH control.

### *$SO_2$ Removal Efficiency*

A survey of the seven companies with experience in full-scale tail-end wet scrubbing shows that most companies will provide $SO_2$ removal guarantees varying from 70 to 90 percent, or as required to meet EPA standards.

### *Advantages and Disadvantages*

The advantages of lime and limestone systems are as follows:

1. Relatively low capital and operating costs;
2. Potentially high $SO_2$ removal efficiencies;
3. Ability to simultaneously remove both $SO_2$ and particulates;
4. Most fully characterized of FGD systems.

The disadvantages are:

1. The requirement to dispose of large quantities of waste sludge in an environmentally acceptable manner;
2. The tendency toward chemical scaling, plugging, and erosion problems if not carefully designed and operated.

### *Performance and Reliability*

Operating experience at the Mitsui, Shawnee, and Louisville Gas & Electric scrubbers indicates that lime/limestone scrubbers which have the alkali added to the scrubber circuit can be made to operate reliably. The Mitsui unit in particular has demonstrated the viability of this technology via reliable operations extending over 1½ years. However, experience at other facilities indicates that reliability can be a problem if the systems are not carefully desgined and operated. Also, the boiler injection mode has proven to be a troublesome configuration (Union Electric and Kansas Power & Lights units) which is prone to potentially serious operating problems.

## 2. *DOUBLE ALKALI FLUE GAS DESULFURIZATION PROCESS*

### *Process* (see fig. A-2)

The many double alkali process variations involve the scrubbing of flue gases with a clear liquor containing dissolved sodium or ammonium salts, followed by treatment of the spent liquor with lime or limestone in a reaction producing a throwaway sludge for disposal and regenerated alkali liquor for scrubbing. Typical reactions occurring in the scrubber and reaction tank, respectively, are as follows for a sodium-based system:

$$Na_2SO_3 + SO_2 + H_2O \rightarrow 2NaHSO_3; \qquad (3)$$

$$2NaHSO_3 + Ca(OH)_2 \rightarrow Na_2SO_3 + 3/2H_2O + CaSO_3 \cdot \tfrac{1}{2}H_2O \qquad (4)$$

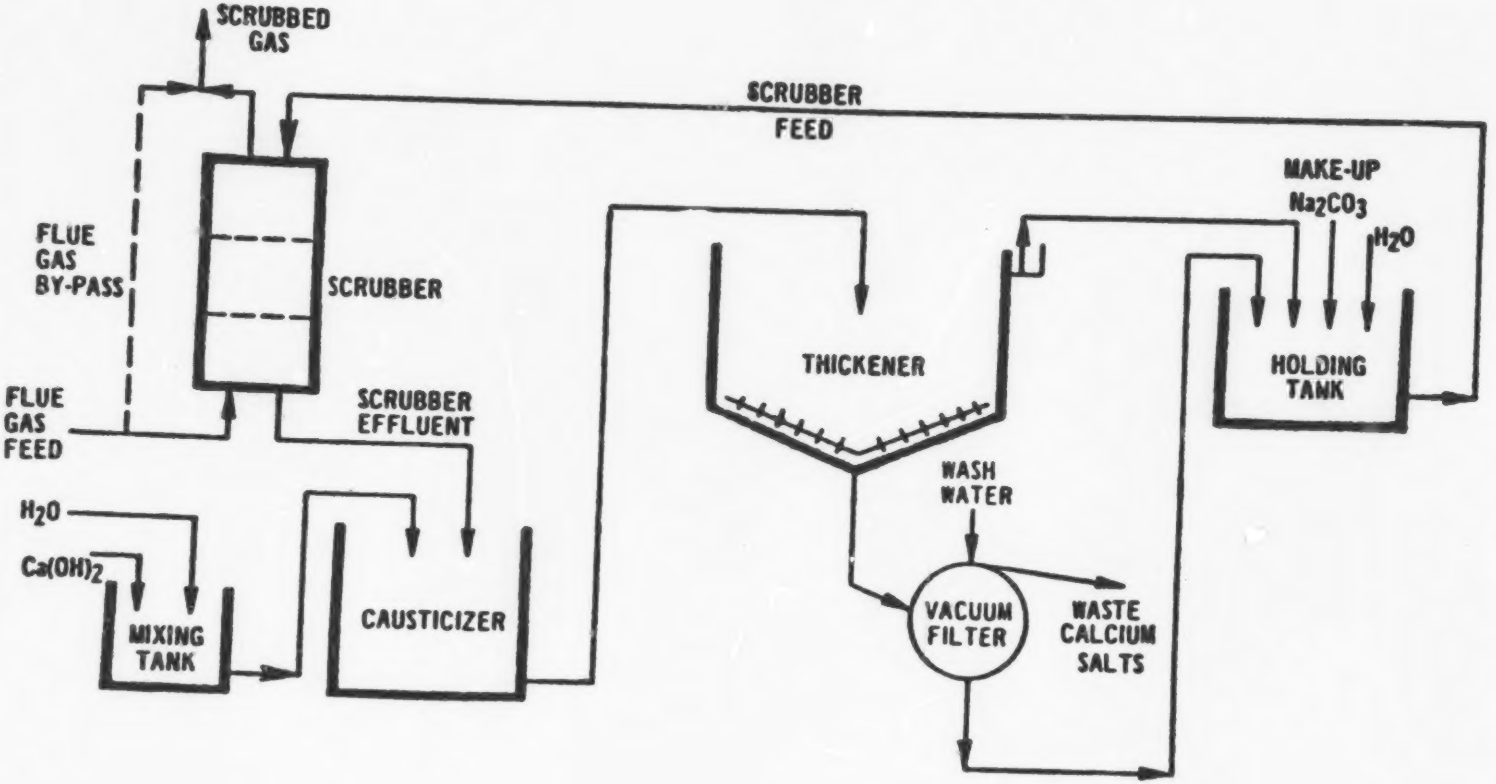
### *History and Experience*

In the United States, primary developmental attention has been placed on sodium-based double alkali systems using lime for regeneration. Important pilot plant work has been performed by General Motors, FMC Corporation, Envirotech, A.D. Little/Combustion Equipment Associates, and Chemico.

Double alkali systems have been intensively developed in Japan, with limestone used as the input alkali and gypsum produced as a saleable product. Showa Denko has recently started up a 156 MW system on an oil-fired power plant. Two additional 150 MW units, on oil-fired oilers using Kawasaki/Kureha technology, are under construction and will start up in mid-1974.

### *$SO_2$ Removal Efficiency*

Based on pilot scale results to date, $SO_2$ removal efficiences between 90 and 99 percent are achievable at reasonable costs.



SCRUBBED GAS
SCRUBBER FEED
FLUE GAS BY-PASS
SCRUBBER
FLUE GAS FEED
SCRUBBER EFFLUENT
$H_2O$
$Ca(OH)_2$
MIXING TANK
CAUSTICIZER
THICKENER
WASH WATER
VACUUM FILTER
WASTE CALCIUM SALTS
MAKE-UP
$Na_2CO_3$
$H_2O$
HOLDING TANK

FIGURE A-2. DOUBLE ALKALI PROCESS VARIATION – SODIUM SULFITE SCRUBBING WITH REGENERATION OF THE SULFITE WITH LIME

*Advantages and Disadvantages*

The advantages of double alkali systems include:

1. Relatively low capital and operating costs;
2. Very high $SO_2$ removal efficiencies;
3. Use of clear solution scrubbing minimizes solids buildup and erosion problems offering potential for high reliability;
4. Ability to simultaneously remove $SO_2$ and particulates.

Disadvantages include:

1. Requirement to dispose of large quantities of waste sludge in an environmentally acceptable manner;
2. Design complexities necessary to deal with the following problems:
    a. Necessity to prevent excessive purge of $Na_2SO_4$ produced as a result of oxidation ($Na_2SO_4$ is difficult to regenerate),
    b. Necessity to avoid saturation of the clear scrubbing liquor with calcium sulfate which could lead to scaling problems.

*Performance and Reliability Summary*

Based on pilot scale operating experience at General Motors, FMC Corporation, Envirotech, and A.D. Little in the United States, and at Kureha and Showa Denko in Japan, double alkali systems offer potential for extremely high (>90 percent) $SO_2$ removal with high reliability.

## B. SALEABLE PRODUCT FGD SYSTEMS

### 1. *MAGNESIUM OXIDE $SO_2$ SCRUBBING PROCESS*

*Process* (see fig. A-3)

Chemico's MgO process utilizes an aqueous slurry of magnesium oxide, magnesium sulfite, and magnesium sulfate to scrub $SO_2$ from flue gas streams. The major reaction involves the formation of additional magnesium sulfite through combination of $SO_2$ and magnesium oxide. Magnesium sulfite removed from the scrubber loop is dried and subsequently calcined to drive off $SO_2$ and to regenerate active MgO for return to the scrubber loop. The regeneration can be accomplished either at the power plant site, or at some remote location since the magnesium sulfite and magnesium oxide are stable solids capable of being shipped. The $SO_2$ generated in the calcining operation can be converted to high grade sulfuric acid or to elemental sulfur by provision of the appropriate equipment.

*History and Experience*

In June 1970, the EPA entered into an agreement with Chemico, Boston Edison Company, and Essex Chemical Company for construction and operation of the Chemico magnesia slurry process on the No. 6 boiler at Boston Edison's Mystic Station in Everett, Massachusetts. The installation on this 155 MW oil-fired boiler was completed in May 1972. After completion of the system, numerous operational problems developed requiring equipment modifications and investigations directed toward solving the problems. Most of the problems were of a materials handling nature, resulting from the character of the solids generated in the scrubber loop. Most of these problems have been satisfactorily solved as demonstrated by the high availability factor for June and July 1973, and more recently since late February 1974. Some equipment modifications are continuing to improve operability.

An additional MgO system has recently been started (September 1973) at Potomac Electric Power Company's Dickerson Station. This unit will handle half the flue gas from a 195 MW coal-fired boiler. The system is designed to accommodate flue gas from the burning of 3.0 percent sulfur coal. Very early results are encouraging. An additional coal-fired application, designed and engineered by United Engineers & Constructors, is nearing completion at Philadelphia Electric's Eddystone Station. This unit will handle the equivalent of 120 MW with 2.5 percent sulfur coal fuel.

*Advantages and Disadvantages*

The major advantages of the MgO process are summarized as follows:

1. Sulfur can be removed as high grade acid or elemental sulfur depending upon equipment provided for regeneration.
2. Regeneration can be accomplished at a location quite distant from the power plant (for instance, at an existing sulfur acid plant) thus permitting the use of a central regeneration facility servicing several flue gas cleaning locations.
3. By maintaining adequate inventories of MgO, extended outages of the regeneration facility can be tolerated without interruption of the pollution control facility.
4. Process reliability has benefited from the modifications and investigations at the Boston Edison site and will continue to improve as the two additional systems, Potomac and Philadelphia, become fully operational in the near future.

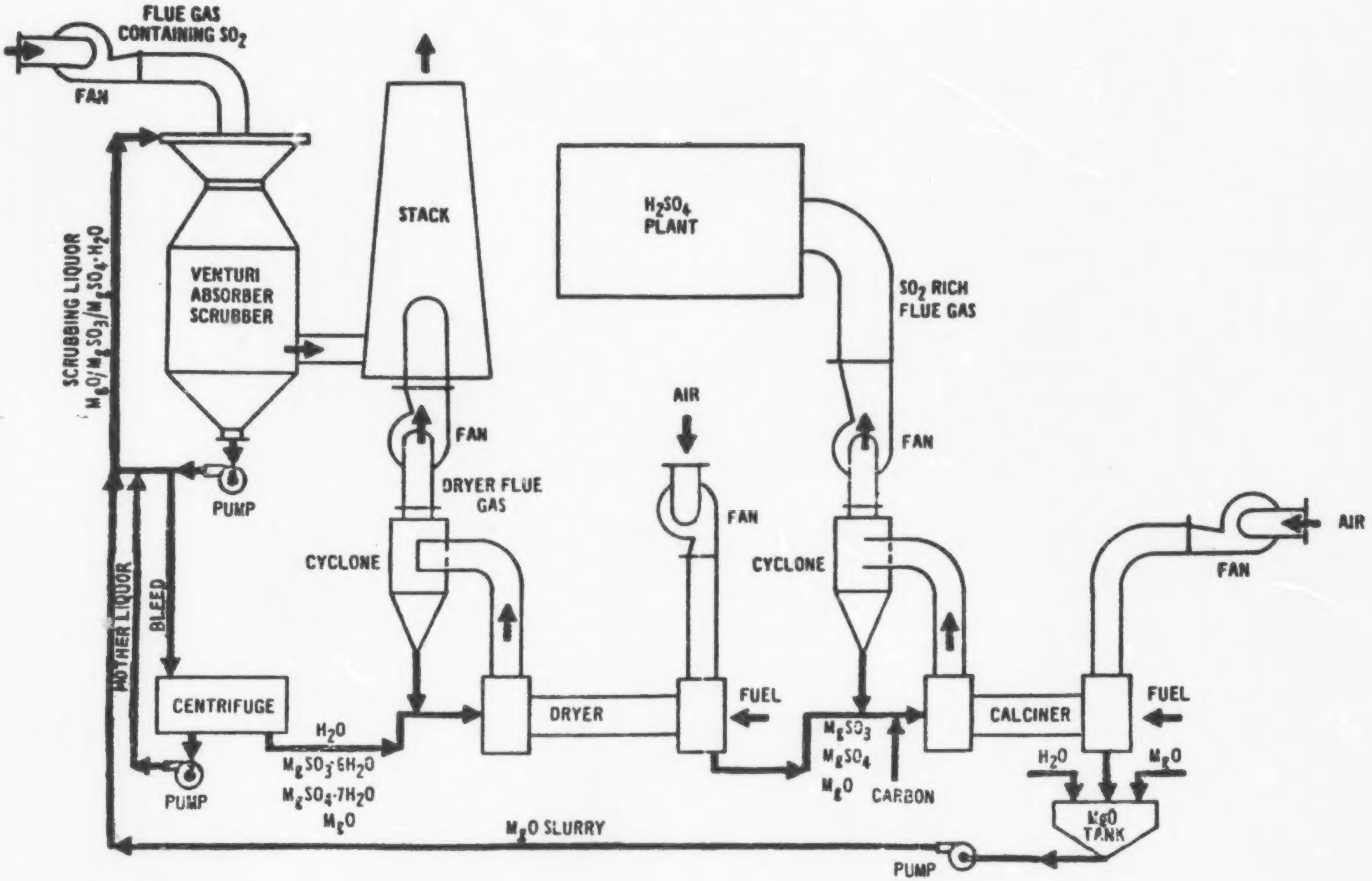

FLUE GAS CONTAINING $SO_2$
FAN
SCRUBBING LIQUOR
$MgO/MgSO_3/MgSO_4 \cdot H_2O$
VENTURI ABSORBER SCRUBBER
PUMP
MOTHER LIQUOR
BLEED
CENTRIFUGE
PUMP
$H_2O$
$MgSO_3 \cdot 6H_2O$
$MgSO_4 \cdot 7H_2O$
MgO
STACK
FAN
DRYER FLUE GAS
CYCLONE
DRYER
AIR
FAN
FUEL
$H_2SO_4$ PLANT
$SO_2$ RICH FLUE GAS
FAN
CYCLONE
$MgSO_3$
$MgSO_4$
MgO
CARBON
CALCINER
FUEL
$H_2O$
MgO
MgO TANK
AIR
FAN
MgO SLURRY
PUMP

FIGURE A-3. MgO SLURRY PROCESS FOR FLUE GAS FREE OF PARTICULATE MATTER

The major disadvantage of the process is the lack of both reliable long-term operating experience and experience with a coal-fired system.

### *$SO_2$ Removal Efficiency*

The MgO process is capable of achieving 90 percent $SO_2$ removal over a wide range of inlet $SO_2$ concentrations.

### *Performance and Reliability*

To date, the experience at Boston Edison has established the capability of the process to consistently achieve 90 percent $SO_2$ removal. Also, the high turndown capability and reliable operation of the venturi scrubber configuration used at Boston Edison have been established. The reliability of the entire MgO process has not been established through long-term operation; however, the onstream time at Boston Edison has improved with the various modifications, as indicated by an 85 to 90 percent availability for June and July 1973. Modifications to improve system reliability are continuing.

## 2. *WELLMAN-LORD FGD PROCESS*

### *Process* (see fig. A-4)

The Wellman-Lord (W-L) $SO_2$ recovery process utilizes a sodium sulfite-sodium bisulfite solution to absorb $SO_2$ from flue gas. The spent absorbent, rich in bisulfite, is processed in a steam-heated evaporator, regenerating active sodium sulfite and producing a stream of $SO_2$ for further processing. The process, depicted in simplest for is:

$$\textit{Absorption:}\quad SO_2 + Na_2SO_3 + H_2O \rightarrow 2NaHSO_3 \tag{5}$$

$$\textit{Regeneration:}\quad 2NaHSO_3 \underset{heat}{\rightarrow} Na_2SO_3 \downarrow + SO_2 \uparrow + H_2O \uparrow \tag{6}$$

Inactive sodium sulfate is formed by three mechanisms: $SO_3$ absorption, disproportionation, and sulfite oxidation. Sodium sulfate must be purged from the system in order to maintain adequate levels of active sulfite in the absorber/evaporator loop.

### *History and Experience*

Patents for the W-L process were filed in 1966, and patent rights are currently held by Davy Powergas Inc. Lakeland, Florida. Two Japanese firms are licensed to market the system. There are currently seven operating plants in the United States and Japan, including two oil-fired boiler applications. In addition, more than 10 systems are on order including 5 additional boiler applications. The earliest operating W-L system is the sulfuric acid plant applications at Paulsboro, New Jersey, which has been operating since July 1970. The earliest boiler application is the industrial boiler (oil-fired) at Japan Synthetic Rubber (JSR) Company, Chiba, Japan. This unit, which is equivalent to 75 MW, has been operating successfully since August 1971 and has an excellent onstream factor of 97 percent since that time. It is worthy of note that both JSR and Olin Corporation (owner of the Paulsboro system) have ordered additional systems, thus attesting to the satisfactory performance of the W-L system. Standard Oil of California purchased a W-L system for Claus Plant tailgas cleanup which was started in June 1972. As a result of the performance of the system, Standard Oil has ordered three additional W-L systems for other locations.

The largest unit in operation is on a 220 MW oil-fired utility boiler operated by Chubu Electric in Japan. This unit has been operating since May 1973 with particular success in minimizing the quantity of sulfate purge.

The EPA has undertaken the demonstration of the W-L system on a 115 MW coal-fired boiler at Northern Indiana Public Service Company's D.H. Mitchell Station in Gary, Indiana. The capital cost of the system is being cost-shared on an equal basis with the using utility, NIPSCO; the demonstration year is scheduled to start in the fall of 1975. In the case of the EPA/NIPSCO demonstration unit, the W-L system will be mated with the Allied Chemical $SO_2$ reduction process to produce elemental sulfur. Allied Chemical will operate and maintain the W-L/Allied system under contract with NIPSCO.

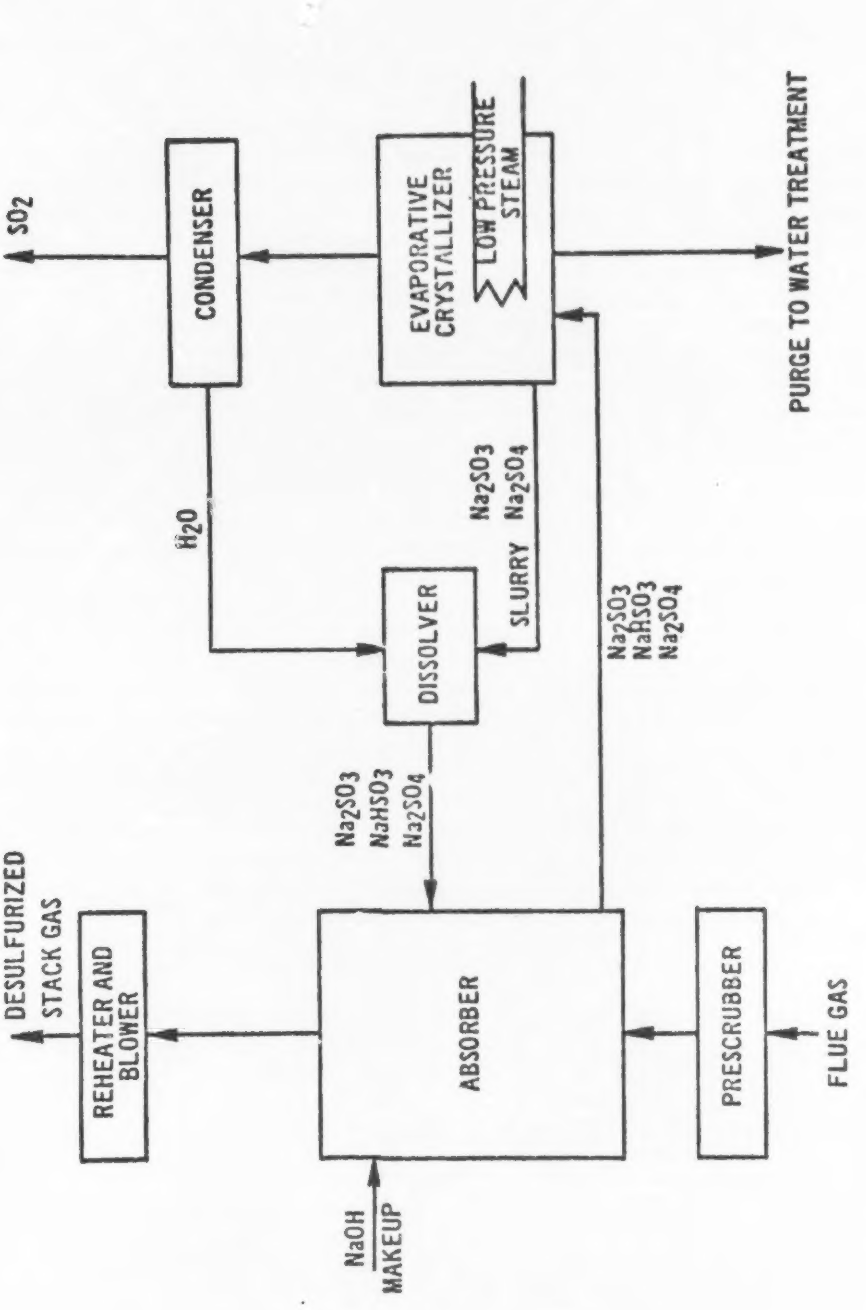
### *$SO_2$ Removal Efficiency*

W-L systems have obtained greater than 90 $SO_2$ removal efficiency for commercial systems operating for long periods of time.

### *Advantages and Disadvantages*

The W-L process has several major advantages as follows:

1. Simplicity and reliability of the various unit operations involved;
2. Ability to produce elemental sulfur or high grade sulfuric acid;
3. High efficiency $SO_2$ removal when required (95 percent or better);
4. Surge capacity before and after the absorber to handle flue gas surges and to enhance system reliability;
5. Many applications and considerable operating experience which provide confidence for success in future applications.



DESULFURIZED STACK GAS
REHEATER AND BLOWER
NaOH MAKEUP
ABSORBER
PRESCRUBBER
FLUE GAS
$Na_2SO_3$ $NaHSO_3$ $Na_2SO_4$
DISSOLVER
$H_2O$
SLURRY
$Na_2SO_3$ $Na_2SO_4$
$Na_2SO_3$ $NaHSO_3$ $Na_2SO_4$
CONDENSER
$SO_2$
EVAPORATIVE CRYSTALLIZER
LOW PRESSURE STEAM
PURGE TO WATER TREATMENT

FIGURE A-4. WELLMAN–LORD PROCESS SCHEMATIC

Major process difficulties and disadvantages are:

1. Need to sell or dispose of a quantity of purge solids (sodium sulfate and other sodium salts);
2. High energy demand results in derating of power station (3 to 6 percent);
3. No coal-fired applications in operation.

*Performance and Reliability Summary*

Operating W-L systems have consistently achieved $SO_2$ removal efficiencies in excess of 90 percent for a wide range of $SO_2$ inlet concentrations. The most significant development efforts to date have been in the areas of reduce capital costs and minimization of purge requirement. As a result of process modifications, which have now been proven in various operating systems, purge requirements of 5 percent or less of inlet sulfur can be anticipated.

Both the inherent high reliability of the equipment involved in the W-L process and the actual operating experience (97 percent onstream factor over a 2-year period at the JSR boiler installation) point to an availability in excess of 95 percent. The high system reliability would be enhanced in large-scale systems due to the opportunity for greater use of equipment spares and partial systems operation as a result of multitrain configuration. The current practice of providing absorbent surge capacity permits short-term shutdown of the regeneration process without interrruption of the scrubbing capability.

## 3. CAT-OX FGD PROCESS

*Process* (see fig. A-5)

The Monsanto Cat-Ox Process utilizes catalytic oxidation to convert most of the $SO_2$ present in the flue gas stream to $SO_3$ for subsequent removal by an acid-absorbing tower (followed by a fiber-packed mist eliminator to remove $H_2SO_4$ mist). For retrofit or "reheat" applications, the flue gases emerging from the boiler are passed through a high efficiency (99.6 percent) precipitator and then heated ("reheated") to 850°F (454°C) as preparation for the catalytic oxidation step. The strength of acid produced in the absorbing tower is ~80 percent, which is primarily suitable for use in fertilization production.

*History and Experience*

The Cat-Ox process was piloted on a 15 MW scale for 2 years, commencing in August 1967. Based on the successful pilot operation, agreement was reached in June 1970 between EPA, Illinois Power, and Monsanto to install and operate a demonstration Cat-Ox system on a 110 MW coal-fired boiler at Illinois Power's Wood River Station at East Alton, Illinois. Construction of the system was completed in July 1972; after a considerable debugging period, the system was acceptance-tested in July 1973. Because of the present scarcity of natural gas, the reheat burners must now be modified to provide the capability for 100 percent firing on fuel oil. This has occasioned another delay which will preclude commencement of the yearlong demonstration program until approximately May 1974.

*$SO_2$ Removal Efficiency*

During the acceptance test, the Illinois Power System achieved emission control guarantees of 85 percent $SO_2$ removal and 99 percent particulate removal. Removal efficiencies of greater than 90 percent are achievable with this technology for many applications.

*Advantages and Disadvantages*

The Cat-Ox process advantages are as follows:

1. The generation of a product which, in certain limited locations, can be disposed of by sale;
2. Operating costs are relatively low;
3. $SO_2$ removal is consistently 85 percent or better over a wide range of $SO_2$ input concentrations.

Major process difficulties and disadvantages are:

1. Cat-Ox must be used near an appropriate acid user, and dilute acid can be difficult to market in large quantities in some locations;
2. Capital costs are high;
3. Reliability and maintenance costs are not currently established due to lack of operating experience.

*Performance and Reliability Summary*

Work to date at the Wood River demonstration site has established the capability of the system to achieve 85 percent $SO_2$ removal; however, no information is available regarding system reliability or any possible performance degradation as a function of operating time. It should be noted that the process and equipment are similar to the standard contact acid process, and that major reliability uncertainties associated with the process center around the ability of the precipitator to provide adequate particulate removal to protect the catalyst bed.

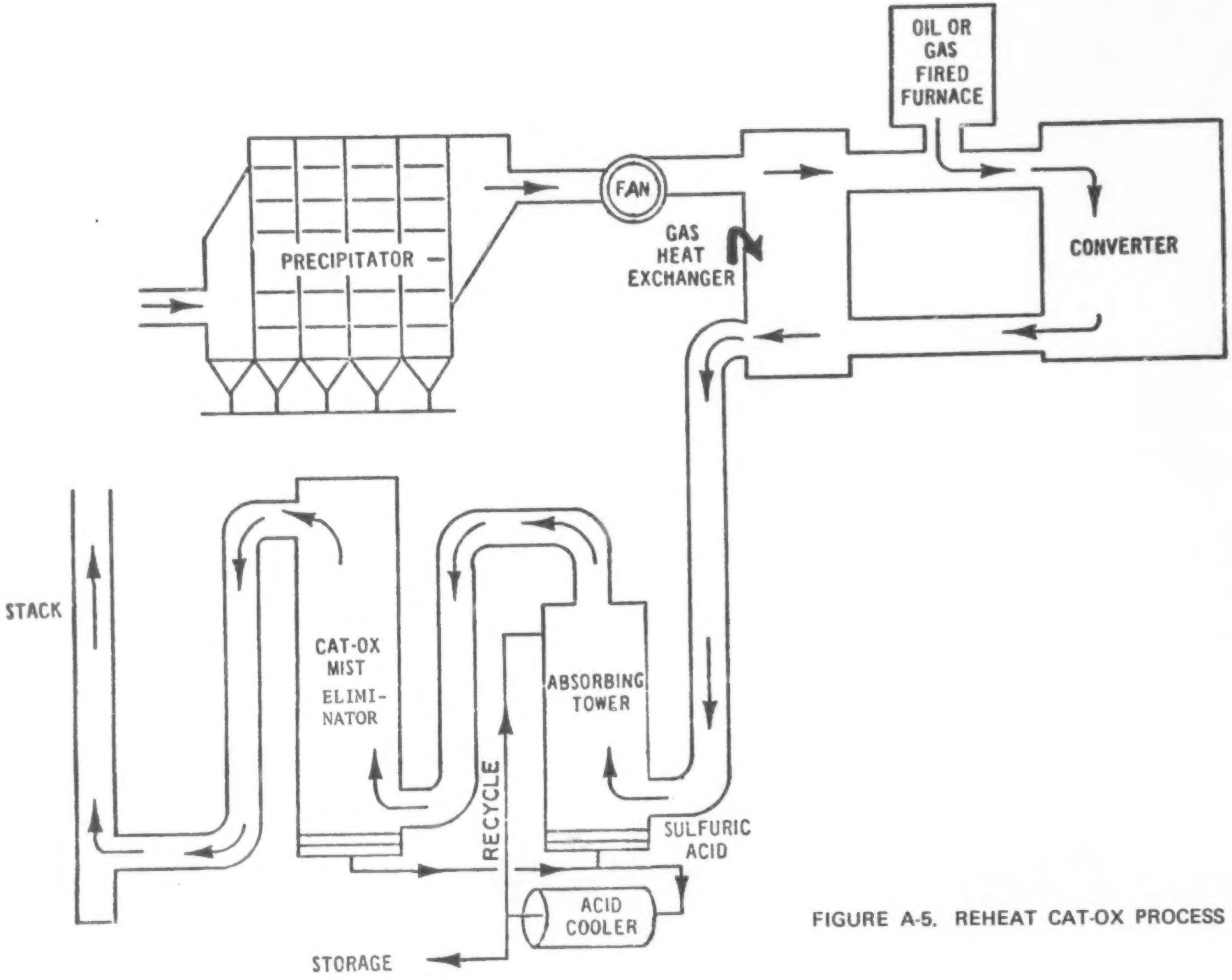

OIL OR
GAS
FIRED
FURNACE
PRECIPITATOR
FAN
GAS
HEAT
EXCHANGER
CONVERTER
STACK
CAT-OX
MIST
ELIMI-
NATOR
ABSORBING
TOWER
RECYCLE
SULFURIC
ACID
ACID
COOLER
STORAGE

FIGURE A-5. REHEAT CAT-OX PROCESS

# APPENDIX B
# OPERATING AND PLANNED FGD SYSTEMS ON U.S. POWER PLANTS AS OF SEPTEMBER 1973

TABLE B-1. OPERATING AND PLANNED FGD UNITS ON
U. S. POWER PLANTS AS OF SEPTEMBER 1973--LIMESTONE SCRUBBING

| Utility Company Power Station | New or Retrofit | Size of FGD Unit, Mw | Process Vendor | Fuel, Sulfur Content, % | Status (Start-Up Date) |
|---|---|---|---|---|---|
| Commonwealth Edison Will County No. 1 | R | 156 | B&W | Coal, 3.5 | Operational (Feb. 1972) |
| Kansas City Power & Light, Hawthorn No. 4 | R | 100 | CE | Coal, 3.5 | Operational (Aug. 1972) |
| Kansas City Power & Light, LaCygne Sta. | N | 820 | B&W | Coal, 5 | Operational (June 1973) |
| Arizona Public Service Cholla Station | R | 115 | Research Cottrell | Coal, 0.4 - 1.0 | Operational (Dec. 1973) |
| Detroit Edison St. Clair No. 6 | R | 180 | Peabody Engineering | Coal, 3.7 | Under construction (Mid-1974) |
| Southern California Edison (operating agent) Mohave Sta. | R | 160 | UOP | Coal, 0.5 - 0.8 | Under construction (Mid-1974) |
| TVA Widow's Creek No. 8 | R | 550 | TVA | Coal, 3.7 | Under construction (Oct. 1975) |
| Northern States Power Sherburne County No.1 | N | 680 | CE | Coal, 1 | Under construction (May 1976) |
| Public Service of Indiana, Gibson Sta. | N | 650 | CE | Coal, 1.5 | Planned (1976) |
| Northern States Power Sherburne County No. 2 | N | 680 | CE | Coal, 1 | Planned (May 1977) |

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TABLE B-2. OPERATING AND PLANNED FGD UNITS ON
U. S. POWER PLANTS AS OF SEPTEMBER 1973--LIME SCRUBBING

| Utility Company Power Station | New or Retrofit | Size of FGD Unit, Mw | Process Vendor | Fuel, Sulfur Content, % | Status (Start-Up Date) |
|---|---|---|---|---|---|
| Union Electric Co. Meramec No. 2 | R | 140 | CE | Coal, 3 | Abandoned (Sept. 1968) |
| Kansas Power & Light Lawrence No. 4 | R | 125 | CE | Coal, 3.5 | Operational (Dec. 1968) |
| Kansas Power & Light Lawrence No. 5 | N | 430 | CE | Coal, 3.5 | Operational (Nov. 1971) |
| Kansas City Power & Light, Hawthorn No. 3 | R | 100 | CE | Coal, 3.5 | Operational (Nov. 1972) |
| Louisville Gas & Electric Paddy's Run No. 6 | R | 70 | CE | Coal, 3 | Operational (April 1973) |
| Duquesne Light Co. Phillips Station | R | 100 | Chemico | Coal, 2 | Operational (March 1974) |
| Southern California Edison (operating agent) Mohave Sta. | R | 160 | SCE/Stearns-Roger | Coal, 0.5 - 0.8 | Under construction (Mid-1974) |
| Ohio Edison/Mansfield Sta. (2 units) | N | 1650 | Chemico | Coal, 4.3 | Under construction (Early 1975) |
| Montana Power Colstrip No. 1 & 2 | N | 720 | CEA | Coal, 0.8 | Under construction (May 1975) |
| Columbus & Southern Conesville No. 5 & 6 | N | 750 | Not selected | | Planned (1976) |

TABLE B-3. OPERATING AND PLANNED FGD UNITS ON
U. S. POWER PLANTS AS OF SEPTEMBER 1973--L/LS NOT SELECTED

| Utility Company Power Station | New or Retrofit | Size of FGD Unit, Mw | Process Vendor | Fuel, Sulfur Content, % | Status (Start-Up Date) |
|---|---|---|---|---|---|
| Salt River Project Navajo No. 1 | N | 750 | Not selected | Coal, 0.5-0.8 | Construction start Nov. 1974 (Mar.'76) |
| Salt River Project Navajo No. 2 | N | 750 | Not selected | Coal, 0.5-0.8 | Construction start Oct. 1975 (Oct.'76) |
| Arizona Public Ser. Four Corners No. 1 | R | 175 | Not selected | Coal, 0.75 | Construction start Oct. 1975 (Oct.'76) |
| Arizona Public Ser. Four Corners No. 2 | R | 175 | Not selected | Coal, 0.75 | Construction start Nov. 1975 (Dec.'76) |
| Southern California Edison (operating agent) Mohave No. 1&2 | R | 1180 | Not selected | Coal, 0.5-0.8 | Planned (Dec. 1976) |
| Arizona Public Ser. Four Corners No. 3 | R | 229 | Not selected | Coal, 0.75 | Construction start June 1976 (Mar.'77) |
| Salt River Project Navajo No. 3 | N | 750 | Not selected | Coal, 0.5-0.8 | Construction start Mar. 1976 (Mar.'77) |
| Arizona Public Ser. Four Corners No. 4 | R | 800 | Not selected | Coal, 0.75 | Construction start Sept. 1975 (Apr.'77) |
| Arizona Public Ser. Four Corners No. 5 | R | 800 | Not selected | Coal, 0.75 | Construction start Nov. 1976 (June '77) |

TABLE B-4. OPERATING AND PLANNED FGD UNITS ON
U. S. POWER PLANTS AS OF SEPTEMBER 1973-- Mag-Ox SCRUBBING

| Utility Company Power Station | New or Retrofit | Size of FGD Unit, Mw | Process Vendor | Fuel, Sulfur Content, % | Status (Start-Up Date) |
|---|---|---|---|---|---|
| Boston Edison Mystic No. 6 | R | 150 | Chemico | Oil, 2.5 | Operational (April 1972) |
| Potomac Electric & Power Dickerson No. 3 | R | 100 | Chemico | Coal, 2 | Operational (Sept. 1973) |
| Philadelphia Electric Eddystone No. 1 | R | 120 | United Engineers | Coal, 2.5 | Operational (April 1974) |

TABLE B-5. OPERATING AND PLANNED FGD UNITS ON
U. S. POWER PLANTS AS OF SEPTEMBER 1973--OTHER $SO_2$ CONTROL SYSTEMS

| Utility Company Power Station | New or Retrofit | Size of FGD Unit, Mw | Process Vendor | Fuel, Sulfur Content, % | Status (Start-Up Date) |
|---|---|---|---|---|---|
| Catalytic Oxidation (Cat-Ox) | | | | | |
| Illinois Power Co. Wood River No. 4 | R | 110 | Monsanto | Coal, 3.2 | Operational (Oct. 1972) |
| Wellman-Lord | | | | | |
| Northern Indiana Public Service, D.H.Mitchell No.11 | R | 115 | Davy Powergas/ Allied Chemical | Coal, 3.5 | Under construction (Early 1975) |
| Aqueous Sodium Base Scrubbing, Non-Regenerable | | | | | |
| Nevada Power Reid Gardner No. 1&2 | R | 250 | CEA | Coal, 0.5 - 1.0 | Operational (April 1974) |
| Nevada Power Reid Gardner No. 3 | R | 125 | CEA | Coal, 0.5 - 1.0 | Under construction (1975) |

TABLE B-6. OPERATING AND PLANNED FGD UNITS ON
U. S. POWER PLANTS AS OF SEPTEMBER 1973--PROCESS NOT SELECTED

| Utility Company Power Station | New or Retrofit | Size of FGD Unit, Mw | Process Vendor | Fuel, Sulfur Content, % | Status (Start-Up Date) |
|---|---|---|---|---|---|
| Public Service of New Mexico San Juan No. 2 | R | 100 | Not Selected | Coal, 0.8 | Planned (Nov. 1974) |
| Potomac Electric & Power Chalk Point No. 3 | N | 630 | Not Selected | Oil | Planned (1975) |
| Potomac Electric & Power Chalk Point No. 4 | N | 630 | Not Selected | Oil | Planned (1976) |
| Potomac Electric & Power Dickerson No. 4 | N | 800 | Not Selected | Coal, 2 | Planned (1976) |
| Potomac Electric & Power Dickerson No. 5 | N | 800 | Not Selected | Coal, 2 | Planned (1977) |

# APPENDIX C
# SELECTED $SO_2$ CONTROL SYSTEMS ON JAPANESE BOILERS

TABLE C-1. SELECTED $SO_2$ CONTROL SYSTEMS ON JAPANESE BOILERS (September 1973)

| Company and Plant | Process | Size of FGD Unit, Mw | Process Vendor | Fuel | Status (Start-Up Date) |
|---|---|---|---|---|---|
| Japan Synthetic Rubber Chiba Plant | Wellman-Lord | 75 | Davy Powergas (MKK) | Oil | Operational (June 1971) |
| Chubu Electric Yokkaichi Plant | DAP-Mn | 110 | Mitsubishi H.I. | Oil | Operational (Feb. 1972) |
| Mitsui Aluminum Co. Omuta Plant | Lime Scrubbing | 156 | Chemico-Mitsui | Coal | Operational (March 1972) |
| Tokyo Electric Kashima Plant | Carbon Adsorption | 139 | Hitachi Ltd. | Oil | Operational (1972) |
| Fuji Kosan Kainan Plant | Dilute Sulfuric Acid-Gypsum | 52 | Chiyoda | Oil | Operational (Oct. 1972) |
| Chubu Electric Nishinagoya Plant | Wellman-Lord | 220 | Davy Powergas (MKK) | Oil | Operational (May 1973) |
| Showa Denko Chiba Plant | Double Alkali (Limestone) | 156 | Showa Denko | Oil | Operational (Aug. 1973) |
| Chugoku Electric Mizushima | Wet Limestone | 100 | Babcock-Hitachi | Oil | Operational (Nov. 1973) |
| Nihon Synthetic Rubber Yokkaichi Plant | Wellman-Lord | 132 | Davy Powergas (MKK) | Oil | Operational (1973) |

Cont. TABLE C-1. SELECTED $SO_2$ CONTROL SYSTEMS ON JAPANESE BOILERS
(September 1973)

| Company and Plant | Process | Size of FGD Unit (MW) | Process Vendor | Fuel | Status (Start-Up Date) |
|---|---|---|---|---|---|
| Sumitomo Chiba Chem.<br>Chiba Plant | Wellman-Lord | 118 | Davy Powergas<br>(SCEC) | Oil | Operational<br>(1973) |
| Tokyo Electric<br>Yokosuka Plant | Limestone-<br>Gypsum | 130 | Mitsubishi<br>JECCO | Oil | Operational<br>(1974) |
| Tohoku Electric<br>Hachinoe Plant | Lime-Gypsum | 125 | Mitsubishi<br>JECCO | Oil | Under construction<br>(1974) |
| Kansai Electric<br>Amagasaki Plant | Lime-Gypsum | 125 | Mitsubishi<br>JECCO | Oil | Under construction<br>(1974) |
| Kansai Electric<br>Kainan Plant | Lime-Gypsum | 150 | Mitsubishi<br>JECCO | Oil | Under construction<br>(1974) |
| Tohoku Electric<br>Shinsendai Plant | Double-Alkali<br>(Limestone) | 150 | Kureha-Kawasaki | Oil | Operational<br>(Jan. 1974) |
| Shikoku Electric<br>Shintokushima Plant | Double-Alkali<br>(Limestone) | 150 | Kureha-Kawasaki | Oil | Under construction<br>(1974) |
| Hokurika Electric<br>Shinminato Plant | Dilute Sulfuric<br>Acid-Gypsum | 250 | Chiyoda | Oil | Under construction<br>(June 1974) |
| Mitsubishi Petrochem<br>Yokkaichi Plant | Dilute Sulfuric<br>Acid-Gypsum | 234 | Chiyoda | Oil | Under construction<br>(Dec. 1974) |

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## APPENDIX D

## SUMMARY OF OPERATIONAL EXPERIENCE WITH FULL-SCALE FGD UNITS IN THE UNITED STATES AND JAPAN

### A. OPERATIONAL EXPERIENCE ON U.S. INSTALLATIONS

1. *PROCESS*: BOILER INJECTION OF LIMESTONE FOLLOWED BY WET SCRUBBING–THROWAWAY PRODUCT

*Process Supplier*: Combustion Engineering
*Constructor*: Combustion Engineering
*System Location*: Union Electric Company, Meramec Unit 2, St. Louis, Missouri
*Startup Date*: September 1968

*System Description and History of Operation:*

This 140 MW unit, the first full-scale utility $SO_2$ control system in this country, was officially shut down and abandoned in December 1971, approximately 3 years after its inception. This system consisted of two parallel single-stage marble bed absorbers equipped with demisters and reheaters. It was operated with some clarified liquor recycle.

The reason for abandonment was massive plugging and scaling in the entire boiler and $SO_2$ removal system. This recurrent problem barred the company from keeping the generating unit going for long periods. It has been said that the up-and-down operation of the boiler, necessitated by scrubber problems (due to the fact that no provisions for gas bypass had been made), may have compounded the problems by causing moisture to combine with powdery deposits on the boiler tubes to form concrete-like deposits during periods of boiler shutdown. The implication is that boiler plugging might have been at least minimized by equipping the scrubber system with a bypass, thus allowing the boiler to operate independently of the scrubber and less intermittently.

2. *PROCESS*: BOILER INJECTION OF LIMESTONE FOLLOWED BY WET SCRUBBING–THROWAWAY PRODUCT

*Process Supplier*: Combustion Engineering
*Constructor*: Combustion Engineering
*System Location*: Kansas Power and Light, Lawrence, Kansas, Units No. 4 & 5
*Startup Date*: December 1968 and November 1971

*System Description and History of Operation:*

The 125 MW Unit No. 4, started up in December 1968, was one of the first attempts to design, construct, and operate a full-size $SO_2$ scrubber. The limestone injection method was used. Multiple problems occurred during early stages of operation including corrosion, plugging of lines, and problems with reheaters and demisters. These problems did not include scaling. During the first 4 months of 1971, modifications in the original design enabled operation with some reliability.

When the scrubbers for the much larger 420 MW unit No. 5 were started up in November 1971, the pond which was common to both units became overloaded and scaling occurred. Modifications in the design have been made that reduced these chemical problems, but mechanical problems remain. At the present time, the modular arrangement allows cleaning of the scrubbers without shutting the boiler down.

The experience at Lawrence has proved invaluable in making changes for later Combustion Engineering units such as the successful Paddy's Run plant. The Lawrence Station is, however, set in much of its design as a boiler injection system and in the closeness of the demisters to the absorbing bed. It, therefore, cannot fully benefit from the knowledge that has come from the operation of it and other systems.

Kansas Power and Light is able to operate a limestone $SO_2$ scrubbing system, with significant $SO_2$ removal and some reliability, while serving a large utility generating plant. This is a milestone in the development of $SO_2$ control technology.

3. *PROCESS*: LIMESTONE SCRUBBING WITH THROWAWAY PRODUCT

*Process Supplier*: Babcock & Wilcox
*Constructor*: Babcock & Wilcox
*System Location*: Commonwealth Edison Company, Will County Station No. 1, Romeoville, Illinois, near Chicago
*Startup Date*: February 1972

*System Description and History of Operation:*

Will County Unit 1, with scrubber on line, has a net power output of 156 MW. The system consists of two identical, parallel, wet limestone scrubbing trains, each consisting of a venturi for particulate removal, followed in series by a turbulent contact absorber (TCA) for $SO_2$ absorption. Each absorber and scrubber is equipped with its own recirculation tank.

Spent slurrry is bled to a clarifier which produces liquor for recycle via a 5-acre pond and a thickened sludge (underflow) which is pumped to a ready mix concrete truck where additives are mixed with the sludge in an attempt to make the sludge acceptable as landfill.

Since February 1972, the $SO_x$ control systems have operated intermittently. Generally, $SO_x$ removal efficiencies in the 80 to 90 percent range are attainable during periods of operation. Chemistry is not considered to be a problem with the system. Problems which have been responsible for shutdown of the system can be classified as mechanical and were not all attributable to the $SO_x$ control system. Some of the problems attributed to the limestone $SO_x$ control system are: (1) demister and reheater pluggage; (2) wearing and plugging of spray nozzles; (3) reheater vibration and stress corrosion cracking; (4) plugging of slurry lines; (5) "sulfite blinding" of limestone; and (6) scaling. Some of the other reasons cited for system outage are: (1) expansion joint failure; (2) inspection; (3) boiler outage; (4) motor failure; (5) contractor and operator errors; (6) fan trip; (7) water loss to pump glands; (8) fan damper operation; (9) leaks in slurry lines; (10) limestone supply; and (11) booster fan vibration.

Solutions to almost all of the problems do not require development of new technology. Methods of controlling all of the problems specific to limestone wet scrubbing, except for the reheater problems, have been used with some degree of success at Will County. Installation of Inconel alloy reheater tubes is expected to solve the reheater stress corrosion cracking problem. Plugging is avoided by circulation of high-solid content slurries and using adequate liquid/gas ratios. Demister pluggage is minimized by good demister wash techniques. Wear of nozzles is controlled by proper selection of materials of construction.

Between February and December 1972, the two $SO_x$ control trains accumulated in excess of 1,400 and 1,200 hours of operation, respectively. During the outage periods, many improvements and repairs were made. One of the trains has been shut down since April 1973 and presently is being cannibalized to support the other. Commonwealth Edison is attempting to iron out the problems with one train before attempting to operate both simultaneously.

Since March 1974, reliability of the system has improved, with a continuous run of 23 days successfully completed; recent mist eliminator modifications have appeared successful in minimizing plugging problems.

## 4. *PROCESS*: MAGNESIUM OXIDE SCRUBBING WITH THERMAL REGENERATION

| | |
|---|---|
| *Constructor*: | Chemical Construction Corporation |
| *System Location*: | Boston Edison's Mystic Station<br>Everett, Massachusetts |
| *Startup Date*: | April 1972 |

*System Description and History of Operation:*

The MgO system, which has been applied to the 150 MW oil-fired boiler at Boston Edison's Mystic Station, utilizes a venturi scrubber to contact the magnesium oxide magnesium sulfite slurry with boiler flue gases. The solids removed from the scrubber, rich in magnesium sulfite, are dried in a direct-fired rotary dryer. The dried solids are shipped by truck to the Essex Chemical Company contact acid plant in Rumford, Rhode Island, where the magnesium sulfite is calcined. The $SO_2$ produced by the calcining operation is converted to high-grade sulfuric acid and the MgO produced is shipped back to Mystic Station for reuse in the absorber.

In April 1972, the shakedown period began for the Mag-Ox scrubbing system. The venturi scrubber has operated intermittently since then due to mechanical difficulties. During operation, the scrubber has achieved $SO_2$ removal efficiencies in excess of 90 percent with no apparent scrubber-related problems. The major problem has been with the design and operation of the $MgSO_3$ crystal dryer. Redesign of the dryer and a change of fuel to a low viscosity oil appear to be resolving these problems. Other problems with centrifuging the sulfite crystals from the scrubbing liquor and properly calcining the sulfite to regenerate MgO appear to be manageable. An availability factor of about 80 percent was achieved for June and July 1973. After a series of mechanical difficulties during late 1973; the operation since February 1974 has been substantially improved with an availability to the boiler of about 90 percent.

This project is quite important because it will be the first time the individual steps of scrubbing, centrifuging, and calcining have been integrated in the Chemico process. Partially funded by EPA, the project involves not only the scrubber, centrifuge, and dryer at the Boston Edison plant but also the calcining and acid plants at Essex Chemical Company in Rumford, Rhode Island.

The first large-scale coal-fired application was started up in September 1973, at the Dickerson Station of Potomac Electric and Power. Flue gas from approximately 100 MW of the 195 MW of Dickerson Unit 3 will be processed. Since the plant burns coal (3 percent S, 8 percent ash), the scrubbing facility uses one venturi scrubber to remove the particulate and a second scrubber to remove the $SO_2$. Regeneration of MgO for this system will be carried out at the Essex Chemical facility which also serves the Boston Edison system.

5. *PROCESS*: LIMESTONE SCRUBBING WITH THROWAWAY PRODUCT

*Process Supplier*: Research Cottrell
*Utility*: Arizona Public Service
*System Location*: Cholla Power Station No. 1 near Joseph's City, Arizona
*Startup Date*: December 1973

*System Description and History of Operation*:

The FGD System at the Cholla plant was first placed in service on December 15, 1973. The system performed well with about a 92.6 percent availability factor. One of the major problems encountered was corrosion on the outside of the reheater tubes due to acid condensation. This problem was corrected by insulating the ductwork to the reheaters and the installation of a plate ahead of the reheater to divert any acid condensation away from the tubes. Vibrations in the reheater unit were overcome by installation of cross baffles at the gas inlet to the reheater in order to dampen the vibration. Scales and deposits which accumulated around flooded disc scrubber shaft and restricted its rotation were eliminated by modifying the shaft's gland assembly.

6. *PROCESS*: CAT-OX

*Process Supplier*: Monsanto Enviro-Chem Systems, Inc.
*Constructor*: Leonard Construction Company
*System Location*: Illinois Power Company, Wood River Station, East Alton, Illinois
*Startup Date* :

Construction was completed in July 1972, followed by debugging and modification. Acceptance testing was completed in July 1973, with full operation to commence April 1974, following provision of additional modifications.

*System Description and History of Operation:*

The retrofit or "reheat" version of the Cat-Ox system has been installed on the 110 MW coal-fired Wood River No. 4 boiler operated by Illinois Power. The process accepts flue gases from the discharge of a specially provided high efficiency ESP. The gases are heated to 850°F (454°C) by heat exchange with processed gases from the catalytic converter and by supplemental "reheat" burners. The heated gases pass through the fixed bed catalytic converter where $SO_2$ is oxidized to $SO_3$. An acid absorbing tower followed by a fiber acid mist eliminator serve to remove the $SO_3$ prior to discharge of the flue gas to the stack. Product acid is cooled and stored in tanks.

Since completion of the system in July 1972, a series of mechanical difficulties has occurred and modifications or repairs have been effected. During July 1973, an acceptance test was conducted which established the capability of the system to achieve the specified 85 percent $SO_2$ removal while producing an acid product of somewhat greater than the required 77.7 percent concentration. Also, subsequent testing of the high efficiency ESP has established the particulate removal to be sufficient to prevent undue costs for catalyst cleaning. Long-term operation of the system has been delayed until April 1974, due to late delivery of equipment for a planned system modification to permit operation of the reheat burners on fuel oil rather than natural gas.

7. *PROCESS*: LIMESTONE SCRUBBING WITH THROWAWAY PRODUCT

*Process Supplier:* *Combustion Engineering*
*Constructor*: Combustion Engineering
*System Location*: Kansas City Power and Light, Hawthorn Station, Units No. 3 and 4
*Startup Date*: September 1972

*System Description and History of Operation:*

In September 1972, the 100 MW Kansas City Power and Light's Hawthorn Units 3 and 4 started up. Each of the boilers was originally equipped with a boiler injection system and two identical scrubber modules. Unit No. 4 has recently been converted to a tail end system. Before changing to a tail end system, boiler pluggage problems were experienced on Unit No. 4. Unit No. 3 still uses boiler injection and has not experienced boiler pluggage. No problems have been reported with the fans or reheaters on either unit. The remaining problems appear to be primarily with the recirculation system. Modifications similar to those used successfully at Combustion Engineering's scrubber at Louisville Gas and Electric Company's Paddy's Run plant show great promise of overcoming these problems. These include problems with headers and drain pots. Demister pluggage has been a problem but this appears to be solved. Some settling has occurred in the recirculation tank, but this should be corrected with improvements in the agitation of the tank.

The method of ultimate sludge disposal has not been decided upon. Enough land area is available to enable ponding of the sludge for up to 15 years.

This plant is the last plant installed by Combustion Engineering using the limestone injection technique. Removal efficiencies have been between 70 and 80 percent.

8. *PROCESS*: LIMESTONE SCRUBBING WITH THROWAWAY PRODUCT

| | |
|---|---|
| *Process Supplier*: | Babcock and Wilcox |
| *Constructor*: | Babcock and Wilcox |
| *System Location*: | Kansas City Power and Light, LaCygne Station, Unit No. 1 |
| *Startup Date*: | June 1973 |

*System Description and History of Operation:*

The $SO_2$ scrubber at the new 820 MW LaCygne Station started operation in February 1973. The system consists of seven modules. At present, one module is controlling $SO_2$ and particulate with a venturi scrubber and a grid plate scrubber. The limestone is added to the scrubber rather than to the boiler.

The remaining six modules at LaCygne are controlling particulate only, using the venturi section. The grid plate will be installed and limestone added at a future date to control $SO_2$ for the entire plant.

It is too early to accurately assess the reliability of this system, since the boiler is new and has not yet become fully operational. The system is essentially a duplicate of the Will County scrubbing system and has experienced many of the same problems. These include plugging of the demisters, corrosion of the reheater tubes, and other mechanical problems. Like that of Will County, the problems are mechanical in nature and solvable using state-of-the-art methods. It is worthwhile to note that no chemical problems with the scrubber itself have occurred. It appears likely that the system will achieve reliable operation in the near future.

Unlike that of Will County, there are no bypass provisions at LaCygne, and no electrostatic precipitators are installed to collect particulate. The scrubbers are expected to achieve 80 percent control of $SO_2$ as well as satisfactory control of particulate.

A 1-year hold time is available for sludge disposal. Plans beyond that time are uncertain.

9. *PROCESS*: LIME SCRUBBING WITH THROWAWAY PRODUCT

| | |
|---|---|
| *Process Supplier*: | Combustion Engineering |
| *Constructor*: | Combustion Engineering |
| *System Location*: | Louisville Gas and Electric Company, Paddy's Run Station No. 6, Southwest of Louisville |
| *Startup Date*: | April 1973 |

*System Description and History of Operation:*

The $SO_2$ control system installed on this 70 MW electric generating unit consists of two parallel, two-stage marble bed scrubbers installed downstream of an electrostatic precipitator. The scrubber effluent liquor enters a large reaction vessel from which some spent liquor is bled to a thickener followed by a rotary drum filter. A carbide sludge slurry which contains lime as the actve ingredient is fed to the reaction vessel. This absorbent slurry feed is controlled by a measurement of acidity in the system, and changes as necessary with variations in $SO_2$ throughput. $SO_2$ throughput varies with boiler-generator load and with variation of fuel sulfur content.

Between April and August 1973, the unit has accumulated about 1,000 hours of operation. The scrubbers have operated for periods of up to 10-12 days with no problems. $SO_2$ removal has been in the 90 percent range. There has been no evidence of scaling or plugging. Neither erosion nor corrosion has been a problem. There have been some minor mechanical problems. The demisters, two stages of standard chevron, have not been a problem. The demister is washed once every 8 hours with high velocity fresh water at the bottom of the demister. The amount of water used is only that amount needed to make up for normal evaporation and losses with the throwaway sludge. Thus, the system is operated as close to closed loop operation as can be expected.

The limited operation, overall, has not been due to problems for the most part, but due to the lack of operating manpower. During the peak demand periods, when all the Paddy's Run units were on line, LG&E has had to shut the scrubber system down because operating manpower was spread thin and operation of the electric generating equipment took first priority.

## B. OPERATIONAL EXPERIENCE ON JAPANESE INSTALLATIONS

1. *PROCESS*: SOLUBLE SODIUM SCRUBBING WITH THERMAL REGENERATION (WELLMAN-LORD)

| | |
|---|---|
| *Process Supplier*: | Davy Powergas |
| *Constructor*: | Mitsubishi Chemical Machinery Company |

*System Location*: Japan Synthetic Rubber Company (JSR) near Chiba, Japan

*Startup Date*: July 1971

*System Description and History of Operation:*

Successful, reliable operation of the Wellman-Lord $SO_2$ control process at Chiba with essentially 100 percent availability for over 2 years is considered quite significant for the U.S. control situation. This process consistently removes over 90 percent of the $SO_2$ from the exhaust gases of a 75 MW equivalent oil-fired boiler. The load is essentially constant, but the concentration of $SO_2$ varies from 400 to 2,000 ppm.

It appears that the process should be applicable to coal-fired boilers, if flyash removal equipment is installed upstream of the absorber. A Northern Indiana Power Service Company (NIPSCO) unit, partially funded by EPA, will evaluate such systems on a coal-fired boiler. Cost studies indicate that capital and operating costs for a Wellman-Lord system in the United States on a coal-fired boiler are not a great deal higher than those for wet lime/limestone or magnesium oxide scrubbing systems, which are generally considered the least expensive of the flue gas desulfurization systems. The system is commercially offered and guaranteed in the United States.

The major problem with the process is the requirement for a purge to remove contaminants, primarily sodium sulfate ($NA_2SO_4$). Present information indicates about 10 percent of the total incoming sulfur is lost as soluble $Na_2SO_4$ at JSR.

Sodium sulfate is a natural component of sea water but could possibly cause problems in fresh water. This purge has been substantially reduced at the newer installation at Chubu Power and may be further reduced by oxidation retardants research by Sumitomo.

The product of the Wellman-Lord system at JSR is high-purity sulfuric acid. While there may be insufficient markets for the sulfuric acid produced if a large percentage of the plants in the United States used this method, there is probably a market available for the output of several plants, particularly those in eastern and midwesteran industrial states.

Elemental sulfur, which will be produced in the NIPSCO unit, is another potential product which is both storable and potentially saleable; this could ultimately be a more desirable end product, especially for utilities not near a market for sulfuric acid. A disadvantage of elemental sulfur production is the necessity of the use of considerable quantities of reducing agent, such as coke or natural gas.

2. ***PROCESS***: LIME SCRUBBING WITH THROWAWAY PRODUCT

*Process Supplier*: Chemical Construction Company (Chemico)

*Constructor*: Mitsui Miiki Machinery Company

*System Location*: Mitsui Aluminum Company, Omuta Power Station, Japan

*Startup Date*: March 1972

*System Description and History of Operation:*

The scrubber at Mitsui Aluminum's Omuta Power Station began operation on March 29, 1972. The system uses as an absorbant carbide sludge that is chemically identical to lime with regard to $SO_2$ scrubbing. The scrubber serves a coal-fired 156 MW boiler that burns coal equivalent to about 2.5 percent sulfur eastern or midwestern U.S. coal. The scrubber has had several load variations similar to, but less frequent than, typical U.S. boilers. Both $SO_2$ and particulate removal efficiencies have been quite high, 85-90 and 98 percent, respectively. The system has operated at closed-loop conditions for substantial periods of time and is open-loop only occasionally during periods of heavy rain. Flyash concentration into the scrubber is similar to that for typical U.S. retrofits.

The system has operated 24 months, continuously, and essentially trouble free, since startup.

It should be noted that the reliable performance of this system is of real significance to the United States air pollution control program, since the design ground rules for the Japanese unit are quite similar to those of many of our power utilities requiring desulfurization systems. The following are among the areas of commonality: retrofit of existing coal-fired boiler, moderately efficient electrostatic precipitators, installation on moderately large boilers, production of a throwaway product, and availability of lime (calcium hydroxide). The unit takes on additional significance since the system was designed based on U.S. technology (Chemico) and is offered and guaranteed in this country. Two similar units using lime on coal boilers are being constructed in the United States for Duquesne Light Company's Phillips Station and Ohio Edison's Bruce Mansfield Station.

3. ***PROCESS***: SODIUM SCRUBBING WITH THERMAL REGENERATION (WELLMAN-LORD PROCESS)

*Process Supplier*: Davy Powergas, Inc.

*Constructor*: Mitsubishi Chemical Machinery (MKK)

*System Location*: Chubu Electric, Nagoya, Japan
*Startup Date*: May 1973

*System Description and History of Operation:*

The Chubu system is applied to a peaking service, oil-fired utility boiler of 220 MW output. The system utilizes a rectangular sieve tray absorber which has maintained $SO_2$ outlet levels consistently below 150 ppm. Double effect evaporation is utilized for regeneration, and a purge crystallization system is provided to minimize sodium losses through purge. The purge crystals have assayed higher than 85 percent sodium sulfate. The system is designed to treat flue gas from burning of fuel containing 4 percent sulfur and to handle changes in boiler load from 25 to 100 percent. Because of the peaking service application, the system is subjected to frequent weekend shutdowns with no restart problems encountered to date. It is noteworthy that at one point during the recent testing program the boiler fuel was changed from 0.7 to 4 percent sulfur oil instantaneously through boiler operator error. In spite of this many-fold step change in inlet concentration, outlet concentration of $SO_2$ remained below 150 ppm.

15 May 1974

Session V:

# RESEARCH AND DEVELOPMENT NEEDS

Robert P. Hangebrauck
Session Chairman

# TECHNOLOGY NEEDS FOR POLLUTION ABATEMENT IN FOSSIL FUEL CONVERSION PROCESSES

E. M. Magee and H. Shaw*

*Abstract*

*The current status of fossil fuel conversion processes is being evaluated with respect to pollution control and thermal efficiency. An awareness of potential pollution problems will allow the process developers to obviate most of the problems through proper design and construction. Other problems which do not have an apparent solution are also being specified in this study. Early identification of pollution problems will allow research, development, and design work to be carried out in time for implementation in commercial plants. The "technology needs" that have become apparent thus far are reviewed in this paper. The areas that are discussed are:*

- *Dust, Fumes, and Water Runoff from Coal and Shale Storage*
- *Acid Gas Systems*
- *Dirty Process Water Treatment*
- *Trace Elements*
- *Other Technology Needs*
  - *Raw Gas Scrubbing*
  - *Lock Hopper Pressurization*
  - *Noise*
  - *Odors*
  - *Solid Wastes*

## INTRODUCTION

Under contract with the Environmental Protection Agency,† the Exxon Research and Engineering Company is studying the pollution and thermal efficiency aspects of fossil fuel conversion processes. The contract may include all fossil fuels, but work to date has concentrated mainly on coal gasification and liquefaction.

Earlier papers in this symposium have discussed the trace element content of fossil fuels produced or consumed in the United States (ref. 1) and some of the pollution control and thermal efficiency aspects of coal conversion processes (ref. 2). Information gaps and new technology needs in fossil fuel conversion processes are also being defined under this contract and are the subject of the present paper. Although it is necessary to study each conversion process in order to adequately define the technology needs for that process, some areas are general for more than one process and for more than one fossil fuel. These general areas are emphasized in this paper.

## INFORMATION GAPS AND TECHNOLOGY NEEDS

### *Dust, Fumes, and Water Runoff From Coal and Shale Storage*

Large storage piles are, or will be, used in coal conversion, shale conversion, and power generation. In coal conversion plants it is estimated that as much as 1 million tons of coal will be stored in an area of approximately 2 million square feet. The large area and the huge quantity of material involved offer potential for pollution of the surrounding air and water. It is difficult to estimate the quantity of dust dispersed from the storage piles, since this will depend on the physical state of the materials as well as meteorological conditions; but for a rainfall of 6 inches over 24 hours, the water runoff would be 2.5 million pounds per hour.

The dust from the coal piles might be assumed to have the same composition as the bulk coal. This may not be justified for two reasons. First, the dimunition of the coal could lead to enrichment or depletion of certain elements, depending on the physical state of the elemental compounds in the coal. If a particular element is present as microcrystals of its compound, then it could well be enriched in the finer portions of dust. The second reason why dust may not be representative of fresh bulk coal arises because of the ease of surface oxidation of coal when exposed to air (ref. 3, pp. 272-279 and 296-306; ref. 22). That this oxidation is not negligible is illustrated by table 1, which shows the oxygen and carbon dioxide content of the gases in a coal pile at Garrison Dam, North Dakota (ref. 3, p. 299). As can be seen from the table, the oxygen is almost depleted within the coal pile.

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*Authors are with the Government Research Laboratory of Exxon Research and Engineering Company, Linden, New Jersey.

†This work was carried out under contract No. 68-02-0629 with the Environmental Protection Agency.

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Table 1. Oxygen depletion in a coal pile

| Location (months) | $CO_2$, percent | $O_2$, percent |
|---|---|---|
| Top,* (12 mo.) | 10.0 | 0.5 |
| Top,* (20 mo.) | 12.0 | 0.3 |
| Sides, (12 mo.) | 7.3 | 5.4 |
| Sides, (20 mo.) | 11.2 | 2.4 |

*3.5 ft depth.

Table 2. Effects of weathering on physical properties of lignite after 160 days

| Method of storage | Size degration, percent | Friability percent |
|---|---|---|
| Sample at beginning of test | 3.2 | 15.7 |
| Sealed in metal cans | 8.8 | 21.1 |
| Atmospheric | 81.1 | 64.9 |

Table 3. Technology needs in coal and shale storage

- Analyze trace elements in dust in the vicinity of storage piles.
- Analyze organic materials in the air over storage piles.
- Determine runoff water composition from coal or shale piles.

Further evidences of the magnitude of weathering is shown in table 2 (ref. 3, p. 301). Considerable size degradation takes place on weathering, thus increasing the fraction of fines. This could lead to a change in the dust analysis from that of bulk coal.

In addition to dust pollution, air pollution could result from the release of organic materials during oxidation of the coal or shale. This could become especially acute if spontaneous combustion occurs. No information is currently available on the quantity and composition of volatile organic materials present in the air over coal piles.

Of perhaps more concern from the viewpoint of potential pollution is the nature of the liquid effluent (runoff and seepage) from the storage pile. The water analysis might be similar to acid mine water, but this is not for certain due to the increased surface area of the coal or shale and weathering effects. Not only might trace elements be leached from the storage piles, but traces of organic materials might also be removed. The nature of these organic materials cannot be predicted, especially in view of the weathering effects on coal composition.

There are techniques that could be used to contain effluents from storage piles. Silo storage is such a technique; however, since this solution could be expensive, it would be worthwhile to determine the magnitude of the potential problem. The technical effort needed to obtain the appropriate information is indicated in table 3.

### *Acid Gas Systems*

Although acid gas cleanup has been practiced for some time in the petroleum industry, the prospect of deriving synthetic fuels from coal and from shale and the need for releasing cleaner effluents emphasize certain shortcomings of the available technology. Most acid gas removal processes involve absorption of $H_2S$, $CO_2$, COS, etc., in a suitable medium. These processes, including hot carbonate scrubbing, amine scrubbing, low temperature scrubbing with methanol, and others have been available for a number of years. (refs. 4-6 are examples of many publications on the subject, and ref. 7 pertains to recent discussions of several available processes.) Acid gas removal generally follows shift conversion in a gasification plant. The hot gases from the shift converter must be cooled to the absorption temperature which can result in a loss of thermal efficiency of 3 percent or more. The removal of sulfur compounds by reaction with solids is expensive, and the spent solids represent a disposal problem. Thus, incentives exist for research aimed at developing a high temperature system for acid gas removal.

Another deficiency of present day acid gas removal processes, when applied to coal gasification, results from the low ratio of $H_2S$ to $CO_2$ in the effluent streams. This problem arises from the inability of the scrubbers to remove $CO_2$ without contamination from $H_2S$. Thus, the sulfur plant must accommodate the additional volume of $CO_2$. Present practice

Table 4. Technology needs for acid gas removal and treating

- A cheap, efficient, high temperature cleanup system
- Selective removal of most of the $CO_2$ in a stream pure enough to allow venting
- A cheap, clean process to efficiently convert $H_2S$ to sulfur (or nonpolluting compounds of sulfur) that effectively takes care of other trace impurities such as $NH_3$, COS, HCN, hydrocarbons, etc.

usually involves oxidation of $H_2S$ in Claus plants, but this process is difficult to operate with a feed that has a low concentration of $H_2S$. If a process were available for removal of a large fraction of the $CO_2$ at a purity sufficiently high to allow direct venting to the atmosphere, then the sulfur problem would be much easier to manage. A process that could inexpensively remove $H_2S$ from concentrated $CO_2$ would be a good alternative to selective absorption.

Besides the Claus process, a number of other processes are available for oxidizing $H_2S$ to sulfur (e.g., see ref. 8). Some of these are able to treat low concentrations of $H_2S$, but they are expensive and produce water streams that are toxic and difficult to treat (ref. 9). Also, most do not oxidize COS to sulfur.

Claus plants produce a tail gas that contains too high a concentration of gaseous sulfur compounds to allow venting. In many cases, these sulfur compounds must be removed by tail gas treatment. A number of processes are availabe or are under development to accomplish this (refs. 10-12), but they are expensive and have problems such as noxious effluents, inability to handle streams high in $CO_2$ content, or products that are difficult to dispose of.

If the $H_2S$-containing stream is not oxidized in a Claus plant, then another problem may arise. Traces of other components, such as $NH_3$, COS, or light hydrocarbons, may be present along with the $H_2S$. These compounds may go through the sulfur production step unaffected, or they may interfere with the sulfur recovery process.

The technology needs in the area of acid gas removal and treatment are summarized in table 4.

### *Dirty Process Water Treatment*

The area of water effluent treatment needs better definition. One of the chief concerns, as evident in the literature, is the need for phenol removal. Biological oxidation has been investigated as a means for destruction of phenols, and this technique seems to work (e.g., see refs. 13, and 14). As a result, some coal conversion process designs show a biological oxidation unit with dirty water going in and clean water coming out. This oversimplifies the few facts that are available.

Biological oxidation units remove about 90 percent of the biological oxygen demand (BOD) (refs. 13, and 14). Literature is scarce, however, on the types of chemicals that are not removed. The 10 percent BOD that is not removed may represent carcinogenic or other harmful materials. Furthermore, BOD is not the same as total dissolved organics (TDO). The Bureau of Mines has published a list of organic compounds identified in tar from the Synthane process (ref. 15). The compounds included condensed ring aromatics and N-heterocyclics. Some of these compounds may be refractory toward biological removal and hence may escape into the environment. When total water recycle is used, the problem may be magnified; materials that are not removed in the water-treating facility will tend to build up in the recycled water. The chemicals can emerge into the environment from evaporation, with spray from the cooling towers, and in ash sludges.

In addition to potential problems associated with organic materials, problems may also exist with inorganic substances. For example, cyanides and thiocyanates were removed only partially in one installation (ref. 13). Surprisingly, even ammonium ions were not effectively removed. Trace elements would also tend to accumulate in recycled water.

Technology needs in water pollution abatement are summarized in table 5.

### *Trace Elements*

The final major area of concern to be discussed here, and one about which almost nothing is known, is the fate of trace elements in all types of fossil fuel conversion processes. A compilation of available information has been published concerning trace elements in coal, petroleum, and shale (ref. 16), but little information is available concerning the fate of these elements when these fossil fuels are used. The

Table 5. Technology needs in dirty water treatment

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- A study of compounds in water feed to, and effluent from, biological treatment of water from coal gasification and coal and shale liquefaction.
- A study of other techniques for cleaning up this water.

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literature may be summarized briefly. The concentration of various toxic metals was determined for particles of different sizes in particulates in urban air (ref. 17). A study was made of the fate of trace elements during combustion of coal in experimental and industrial furnaces (ref. 18). Mass balances have been made for 34 elements on a coal-fired power station (ref. 19). Finally, the disappearance of a number of elements was studied during coal gasification (ref. 20). The last reference shows that a number of elements disappeared from the ash during gasification, but their fate was not determined. The problems associated with obtaining good material balances point out the critical need for proper sampling and analyses of trace elements.

One potential problem concerning trace elements is the possibility that hazardous elements may be concentrated in some process area and may enter the environment in concentrations much larger than those in the original fossil fuel. When data are available, this aspect of the problem should be analyzed in more detail.

Another possible source of trace element pollution is the use of limestone type materials in flue gas scrubbing. Elements that may exist innocuously as oxides or carbonates in limestones could be released as soluble ions when contacted with $SO_2$ in flue gas scrubbing. Work that will furnish some information in this area will be carried out at Exxon Research and Engineering Company in connection with government-sponsored work on coal combustion in the presence of $SO_2$ acceptor materials. Further work is needed on flue gas scrubbing.

In the area of trace elements, table 6 lists the technology needs that have been identified.

### *Other Technology Needs*

Several other areas where new or improved technology needs are evident will be mentioned briefly:

Table 6. Technology needs in the area of trace elements

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- Material balances of trace elements in all types of fossil fuel conversion processes:
    - Coal and shale liquefaction
    - Coal gasification
    - Coal combustion
    - Flue gas desulfurization
    - Petroleum processing
- Process development to remove trace elements where such processes are needed.

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(1) *Raw Gas Scrubbing*—Large thermal efficiency losses and other technical problems accompany the removal of particulates and tars from coal gasifiers. More efficient techniques are needed.

(2) *Lock Hopper Pressurization*—Dirty gases cannot be used to pressurize lock hoppers, and thermal efficiency losses occur when clean gas is used. Better techniques are needed to feed solids to reactors.

(3) *Noise*—Noise is not usually discussed as a pollutant. It has been found, however, to be a problem in a relatively simple conversion process for producing SNG (ref. 21). The pressurization and depressurization of a large number of lock hoppers could, by itself, cause a noise problem. The problem needs better technical definition.

(4) *Odors*—Like noise, odors can present a problem that is not easily defined. Biological oxidation facilities could be especially obnoxious. Again, there is a need for better technical definition of the problem.

(5) *Solid Wastes*—Assuming that solid waste materials can be changed into all-inorganic effluents, the problem is reduced to that of determining the leachability of trace materials into water supplies and would use the technology discussed under trace elements. Studies on spent oil shale have shown that minor elements are easily leached (ref. 23).

## CONCLUSIONS

Although all technology needs for pollution control have not been discussed, and since many problems are specific to a particular process at a single location, a number of areas have been identified that are of general concern. The most immediate need is for information, since in many cases a decision cannot be made as to whether or not a problem does exist. The technical difficulties in obtaining this information are large and, in many cases, the techniques for obtaining the information (sampling, analytical, etc.) are not fully developed.

The overall problem with defining the pollution aspects of fossil fuel conversion is amplified by the fact that, in most cases, the quantities of pollutants are small when compared with the total plant effluent. Thus, there is a tendency to brush aside the question of pollution with a reference to past experience in some other industry. With some of the new technology that will emerge in the area of fossil fuel conversion, with the high concentration of trace elements and aromatic materials in coal and shale, and with a new awareness of environmental needs, it is possible that minor problems may become major, and that completely new problems may appear. The incentives for research and development in the area of pollution control of fossil fuel conversion processes are certainly larger than ever before.

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# LOW BTU GASIFICATION OF COAL: "WHO NEEDS IT AND HOW CAN IT BE IMPROVED?"

**R. A. Ashworth and B. C. Hsieh***

*Abstract*

*The questionable availability of natural gas and petroleum feedstocks is responsible for various marketing areas opening up for low-Btu coal gas. Commercially available low-Btu coal gasifiers fulfill, to various degrees, the gasifier attributes required for these new market areas. This paper centers attention on two of the areas, industrial heating and advanced gas turbine/steam turbin electric power generation. The desired attributes of low-Btu coal gasification systems are compared with the commercially proved gasifiers to determine further research and development needs. For the industrial heating application, modest research and development is required; however, intensive development work is required for the advanced combined cycle application. A comprehensive low-Btu coal gasification research and development effort for these two market areas will cost an estimated $350 million over the next 15 years.*

## INTRODUCTION

For the first time in this country since the demise of the coal gas producer at the hands of clean and cheap natural gas, low-Btu coal gasification is being seriously considered by private industry. The questionable availability of natural gas and petroleum feedstocks is responsible for various marketing areas opening up for low-Btu coal gas. Low-Btu gasification of coal is a viable alternative for many markets, such as industrial heating, ammonia synthesis, synthesized chemical feedstocks, and electric power generation. For electric power generation, it could replace the scarce fuels now being used in conventional fossil-fueled plants or combined cycle plants. It also has application to future advanced power cycles which will require clean fuels, such as advanced combined cycles, fuel cells, and possible MHD.

We have addressed ourselves to the needs of two of these market areas: industrial heating and advanced gas turbine/steam turbine cycles for electric power generation.

For these two applications, the question is "Can commercially available gasifiers be used to fulfill the needs, especially in light of the greater process and environmental restrictions which may be required?"

## COAL GASIFIER ATTRIBUTES

Figure 1 shows a generalized list of desired requirements for modern day gasifiers as applied to industrial heating and advanced combined cycle power plants. In addition, the manufactured gas must be produced at a total cost that is competitive with other energy forms.

By comparing the attributes of the coal gas producers commercially available today (Lurgi, McDowell-Wellman, Winkler, and Koppers-Totzek) with the desired attributes, attention is focused on the improvements required through advanced research and development. Commercially available gasifiers can now fulfill many industrial heating needs. Therefore, modest research and development effort should suffice for industrial heating applications. However, these gasifiers do not reach the same level of competence in satisfying the more demanding requirements of the combined cycle power systems. Research and development efforts for this application must therefore be more intensive.

Following is a review of the various desired gasifier attributes and the areas where today's gasifiers fall short of our objectives.

### *Nontar Producing Gas*

A coal gas containing tars presents special problems. If tars are present in the coal gas, it is necessary to quickly quench the gas with water as it exits the gasifier in order to prevent related tar condensation problems on heat exchangers and pipeline internals. Consequently, besides tar being somewhat messy to handle, thermal energy is lost in the quench. The fluidized bed Winkler and entrained bed Koppers-Totzek gasifiers can satisfy the desired nontar characteristic because of their high gasifier temperature profile. Fixed beds, because of low design outlet temperature, will always produce tars when bituminous coal is used as a feedstock. However, tar production is not a problem when anthracite coal is used.

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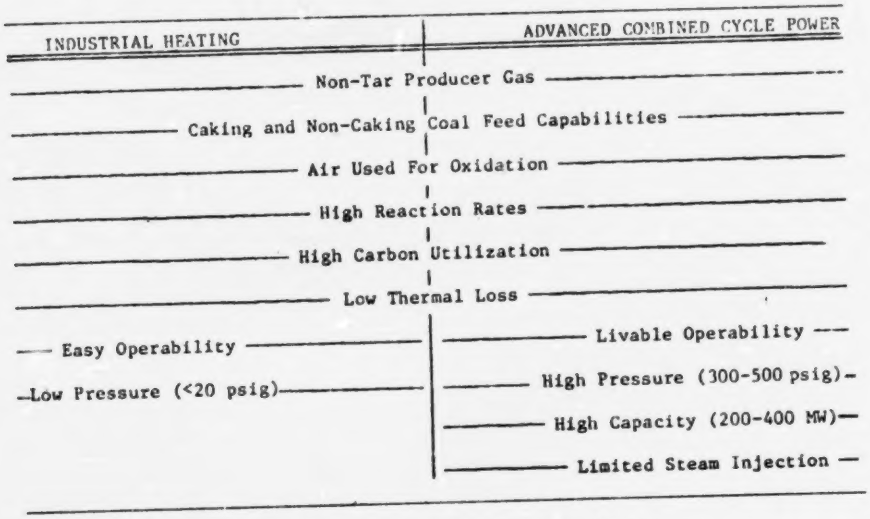
INDUSTRIAL HEATING
ADVANCED COMBINED CYCLE POWER
Non-Tar Producer Gas
Caking and Non-Caking Coal Feed Capabilities
Air Used For Oxidation
High Reaction Rates
High Carbon Utilization
Low Thermal Loss
Easy Operability
Low Pressure (<20 psig)
Livable Operability
High Pressure (300-500 psig)
High Capacity (200-400 MW)
Limited Steam Injection

Figure 1. Desired coal gasifier attributes

To eliminate tar formation from a bituminous coal feedstock, the coal gas must be retained at temperature long enough to allow complete cracking of the tar before it leaves the high temperature region. Consol (ref. 1) has reported that, for a subbituminous coal feedstock, a gasifier temperature of 1,700°F and vapor retention time of 15 seconds produces essentially a nontar gas; however, there is some residual naphthalene. Fifteen seconds for a fairly fast fluidized or entrained bed gasifier is a long retention time. To eliminate tar-producing possibilities, it would seem necessary to maintain gasifier temperatures somewhat higher than 1,700°F. However, the lower the gasifier temperature the better, as it maximizes the Btu content and limits the sensible heat content of the off-gas. Research and development is needed to define the tar-producing parameters for various rank coals in the entrained and fluid beds.

*Caking and Noncaking Coal Feed Capabilities*

Since there are relatively large quantities of caking bituminous coals in the Eastern United States, it would be advantageous to be able to gasify these coals. All three commercial gasifier types (fixed, fluidized, and entrained beds) have demonstrated an ability to gasify caking coals. The fixed bed gasifier has been demonstrated on a pilot scale (ref. 2), but not on a commercial scale. The fluidized bed has been used commercially for gasification of caking coal (ref. 3) at atmospheric pressure, but not at high pressure (ref. 4). Entrained beds (ref. 5) present no problem in handling caking coals.

The need is to prove commercial scale-up of the deep-stirred fixed bed concept so that caking coals can be used directly, without pretreatment, in a fixed bed gasifier. The fluidized bed concept needs to be proved on a commercial pressure gasifier; parameters should also be set for degree of mixing required for various rank coals.

*Air for Oxidation*

The use of air for gasification has been adequately shown on a commercial scale for fixed and fluidized beds; however, this is not the case with an entrained bed, such as the Koppers-Totzek gasifier. For the one-stage entrained bed gasifier, the gasifier temperature must be maintained at a relatively high level in order to provide good carbon conversion efficiency and to keep the ash in a molten state. To yield a combustible Btu content in the gas, air, and steam, preheating may be required. The possibilities of using a one-stage entrained bed gasifier, with air as the oxidizer, should be explored to determine the feasibility of this approach. The use of oxygen for these two low-Btu fuel gas applications is not necessary and is somewhat costly. For the longer term, two-stage entrained bed gasifiers, both high and low pressure, should be developed.

### *High Reaction Rates*

High reaction rates, or high throughput per unit volume, is perhaps the main reason research work is being diverted away from the fixed bed concept to the fluidized and entrained bed types. Since increased temperature, pressure, and gas-solids mixing tends to yield greater throughput per unit volume, the entrained gasifier promises the greatest throughput approach. However, complete entrainment of the coal particles requires delicate control, which can diminish stability of operation. For industrial heating purposes, the fixed and fluidized bed approaches should fulfill most needs. Fluidized and entrained bed gasifiers will compete for advanced combined cycle power application. However, one should not eliminate the fixed bed altogether; it is a good backup.

Though somewhat subject to debate, we can probably generalize and say that: fluidized beds seem to be the best approach to meet intermediate electric power loads and that the entrained beds seem more suitable for base loads.

### *High Carbon Utilization*

To yield the desired high carbon utilization, the two-stage approach applied to entrained and fluid beds may be appropriate. The second stage is provided to effect burnout of the unreacted carbon carried out with the first-stage exit gas and also to increase coal throughput. One stage is sufficient for the fixed bed approach because of the long residence time of coal in the gasifier and the countercurrent flow characteristics of the bed. The one-stage fluid bed Winkler, because of high carbon loss with the ash, yields relatively low carbon utilization. Because of high carbon loss with the ash, yields relatively low carbon utilization. A two-stage atmospheric fluid bed gasifier development would seem to be beneficial for industrial heating applications. It should provide a nontar gas, fair turndown ratio, and livable operating capability. In addition, it could be used a precursor development to the pressurized two-stage fluid bed gasifier for electric power generation. A two-stage atmospheric entrained bed gasifier should be developed to provide good carbon utilization with air as the oxidizer. It would also provide a stepping stone to the pressurized two-stage entrained bed approach for advanced combined cycle power.

### *Low Thermal Loss*

Heat transfer to low temperature heat sinks is a problem in coal gasification. In off-gas cooling, heat exchanger tubes must not exceed much more than 600°F. This is due to the corrosive nature of the $H_2S$-$H_2$ atmosphere. When a coal gasifier is used for electric power generation, some thermal efficiency is lost to these low temperature sinks; the corrosion potential at higher tube wall temperatures being the limiting factor. Necessary water jacketing requirements for entrained bed slagging zones is also a potential heat loss in low temperature heat sinks. Pressure gasification further increases the refractory problems in these hot zones due to increased heat transfer coefficients. Another unsolved problem is the ability of refractory to withstand slag corrosion attack in a reducing atmosphere.

For an advanced gas turbine/steam turbine electric power plant, heat loss to low temperature heat sink in the off-gas can theoretically be minimized by removing the $H_2S$ and particulate hot. The gas cleanup aspects of coal gasification will be considered later. The Btu content of low-Btu gas, with air as the oxidizer, is sensitive to heat loss; as air is added to maintain temperature, the added nitrogen must not only be heated up, but also acts as a diluent, lowering the Btu content of the gas. Heat loss with low-Btu gasification schemes can become quite critical.

### *Operability*

To make coal gasification useful for industrial heating, the gasifier supplied should be easy to operate. When a high pressure gasifier is connected to a gas turbine, the operation is not simple; however, it must be livable!

Fixed bed gasifiers operating at low pressure are the easiest to operate. Turndown ratio is very good. The gasifiers can be banked and restarted with ease (ref. 6). The fluid and entrained beds have lower turndown capabilities, and instrument control is more sophisticated than with the fixed bed. Pressure systems increase possible operational control problems by orders of magnitude for any of the three gasifier types.

### *Pressure*

High pressure coal gasification is not required for most industrial heating needs. However, for combined gas turbine/steam turbine power plants, it is necessary from an overall thermal efficiency standpoint. Less power is required to compress air for a pressure gasifier than to compress an atmospheric gasifier off-gas, because there is less gas, at higher density, to compress. Fixed bed gasifiers (Lurgi) have been commercially demonstrated at 285-430 psig (ref. 7). Fluidized and entrained beds have not been demonstrated at these high-pressure levels. The pressure cases present some difficult problems which must be overcome for both the fluidized and entrained beds. There is also room for improvement in the commercially demonstrated fixed beds.

### *Capacity*

The gasifiers available today are of sufficient capacity per unit to satisfy most industrial heating needs. High capacity units, however, are needed for electric power generation, due to the large heat input requirement. For this reason, we have given values of 200-400 MW capacity per gasifier unit as a goal for developmental effort in the field of electric power generation. As mentioned under "High Reaction Rates," the fluidized and entrained bed approaches yield the higher throughput potential. Fixed bed gasifiers, even high pressure ones, require tremendous land area for an average size power plant. Using the STEAG Power Plant at Lünen (Germany) as a model, an estimated 18 (12-ft diameter) fixed bed gasifiers would be required for a 600 MW combined cycle power plant (ref. 8).

For the short range, atmospheric pressure, air-blown entrained bed gasifiers should be developed for the combined cycle application. For the longer range, pressurized fluid and entrained bed gasifiers should be developed to provide better energy utilization through increased overall thermal efficiency.

### *Steam Injection*

Water added to the gasifier unit is, in the final stage, vented to the atmosphere as a vapor, and much heat is lost due to its high latent heat of vaporization. Steam in excess of that required for gasifier temperature control and gasification reactions should be minimized. A low temperature liquid scrubbing system is another place for water addition, via water evaporation from the scrubbing medium to the coal gas. One of the goals in any process application is to minimize heat loss. It is particularly true for electric power generation where overall thermal efficiency is extremely important. Fixed bed gasifiers require steam, not only for gasification, but also for grate cooling. Lurgi gasifiers with air, require about 0.7 lb steam/lb of subbituminous coal (coal contains 16.5 percent $H_2O$) (ref. 7). Winkler, with oxygen, quotes 0.16 lb steam/lb of 8.6 percent moisture lignite (ref. 9). Koppers-Totzek, with oxygen, quotes 0.16 lb steam/lb of 2 percent moisture subbituminous coal, and 0.29 lb steam/lb of 2 percent moisture Eastern coal (ref. 5).

## MISCELLANEOUS GASIFIER SUPPORT

Besides the major gasification work, much support work would speed up the overall development of the low-Btu coal gasification program. Hot char valves, hot gas valves, hot flanges, hot char recycle, coal feed, and ash removal development programs are very necessary. Metallurgy and refractory development also need intensive side developmental effort.

One item that needs to be more clearly recognized and developed is the instrumentation response and surge capacity required to provide constant fuel gas pressure to the gas turbine, when used in a combined cycle power application. In the pressure Lurgi gasifier combined cycle power plant at Lünen (Germany), no surge capacity is installed other than the vessel capacities themselves (ref. 8). Since many Lurgi gasifiers are required for a moderately sized combined cycle power plant, the gasifiers proper may supply enough surge volume so that added volume will not be necessary (five gasifiers are required for a 170 MW unit). However, this may not be the case with higher throughput per unit volume gasifiers.

Supplemental kinetic and thermodynamic studies may be necessary to aid in scale-up work. Other areas such as economic and operation system studies, environmental impact considerations, and safety analysis for the high pressure cases should also be investigated.

## GASIFIER CLEANUP

Besides the gasifier proper requiring changes to adapt to today's needs, the gas cleanup required for environmental and/or process considerations must be of greater refinement than anything we have needed in the past.

In respect to coal gasification, concern for the environment revolves around two major air pollutants: hydrogen sulfide, which would eventually end up as $SO_2$, and particulate. In addition, if a tar-laden gas is produced, water pollutants such as ammonia, phenols, cyanides, etc., must be addressed. When the coal gas is finally combusted, another air pollutant form, $NO_x$, is produced. These environmental considerations play an important part in any gasification system development.

In figure 2, the environmental and process restrictions are shown for gas effluents as they apply to industrial heating and combined cycle power use. Environmental restrictions are the same in both cases, but added process restrictions are placed on the combined cycle application. Particulate and alkali metal limits are quite restrictive because of erosion and corrosion potential on the gas turbine blades.

At the time of the writing of this paper, the Environmental Protection Agency had not released water effluent standards for coal gasification plants. The assumption we made is that the standards would be similar to those for byproduct coke manufacture, since the contaminant problems are the same. In figure 3, the recently published EPA standards (ref. 12) for new byproduct coke plants are shown.

If cold gas cleanup is specified, the gas cleaning and water contaminant processing aspects should consist only of selection and application of existing commercial processing know-how. Commercial processes are available that should do the job, but necessary demonstrations on coal gas must be performed.

Hot gas cleanup (1,400° to 1,600°F range) is quite another matter. Process development, in this area, is just beginning. As mentioned previously, hot cleanup of hydrogen sulfide and particulate has potential application possibilities in advanced combined cycle power systems. The prime reason for hot cleanup is to provide increased overall thermal efficiencies (ref. 13). The two major gas contaminants, hydrogen sulfide and particulate, must be addressed when hot gas cleanup is considered. For hydrogen sulfide removal, molten salt (ref. 14), iron oxide (ref. 2), and limestone or dolomite (ref. 15) processes are perhaps the front-runners in the research and development stage. To date, however, there is no clear cut choice among the three. The molten salt process must contend with alkali carryover and fly ash contamination of the salt. Iron oxide has shown good potential, but liberates sulfur as $SO_2$, which requires more extensive processing to obtain elemental sulfur than is required with $H_2S$. The limestone and/or dolomite process concepts have been plagued with an inability to regenerate CaS stone back to $CaCO_3$.

Hot particulate removal process development has not been extensive to date, and increased effort is necessary. The hot particulate removal cannot be separated from the hot $H_2S$ removal; they go hand in hand. Particulate removal is critical to the success of hot gas cleanup because of the stringent inlet gas turbine requirements. The panel bed filter (ref. 16) looks very promising for this application.

## RESEARCH AND DEVELOPMENT COST

In scanning the R&D needs for low-Btu gasification, we considered here only the more conventional approaches to coal gasification and coal gas cleanup: fixed, fluidized, and entrained bed gasifiers, and gas cleanup system indepdent of the gasifier proper. Furthermore, we have looked only at the coal gasifier island, which includes the gasifier proper and the gas cleanup systems required to deliver a fuel gas to a boundary limit which would satisfy the fuel requirements of an industrial heating or a gas turbine/steam turbine power plant application. Gas turbine development and molten salt and molten metal gasifiers are not included, but should not be overlooked. These plus advanced power concepts, such as topping and bottoming cycles, magneto-hydrodynamics, fuel cells, etc., should, and are, being investigated.

We will now take a look at estimated cost and time frames needed to accomplish the outlined development. The development has been structured on the premise that the work is important to our national well-being and security. The comprehensive approach proposed here is perhaps costly, but should insure success in the smallest increment of time; time rather than money being considered of prime importance. The problem was approached from two viewpoints, (1) the ambitious, and (2) the conservative. Since time is important, we put emphasis on the ambitious approach; however, the conservative is there for backup. The proposed research work includes the simultaneous development of the conservative and the ambitious approaches in order to more assuredly meet short-range as well as long-range goals.

In figures 4 through 8, we include time and money for the fixed, fluid, and entrained bed gasifiers, miscellaneous gasifier support, and gas cleanup development. The estimates show time and cost required to develop the various concepts to a commercially acceptable state--that is, to develop the process to a point where industry will accept it. The costs and time frames required for the combined cycles cases do not include gas turbine development, or power plant capital and operating cost requirements.

In figure 4, the developmental effort for the fixed bed gasifier is shown to be nominal, approximately

INDUSTRIAL HEATING | ADVANCED COMBINED CYCLE POWER

**$H_2S$**[10]

~1000 ppm $H_2S$ in fuel gas @ 150 Btu/scf to yield 1.2 lb $SO_2$/MM Btu input

**PARTICULATE**

~0.2 gr/scf in fuel gas @ 150 Btu/scf to yield 0.1 lb particles/MM Btu input[10]

— 2-6 MICRON: 0.001 gr/scf in the combustor expansion gas with 6 microns upper limit on particle size[11]

**ALKALI METALS**

— 40 ppb maximum allowable limit in combustor expansion gas[11]

**NOx***

0.2 lb/MM Btu heat input (based on gas)

0.7 lb/MM Btu heat input (based on coal)

* Logically, standard could be based on either gas or coal as fuel.

Figure 2. Environmental and/or process restrictions - gas effluents

| | Maximum for any one day lb/ton coal* | Maximum Average for Any 30 Consecutive days lb/ton coal* |
|---|---|---|
| CYANIDE | 0.0006 | 0.0003 |
| PHENOL | 0.0011 | 0.0006 |
| AMMONIA | 0.0237 | 0.0120 |
| B.O.D.** | 0.0474 | 0.0237 |
| SULFIDE | 0.0009 | 0.0003 |
| OIL & GREASE | 0.0237 | 0.0120 |
| TSS*** | 0.0237 | 0.0120 |
| pH | Within the range of 6.0 to 9.0 | |

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* E.P.A. Standards for by-product coke manufacture. Adjusted from coke to coal by dividing by 0.7.

** Biological oxygen demand.

*** Total suspended solids.

Figure 3. Environmental restrictions - water effluents

|  | To Commercially Demonstrate |  |
|---|---|---|
|  | Estimated Time Frame, Years | Estimated Cost In 1974 Dollars $MM |
| A. Coal Feed Capability* | 4 - 6 | 2.5 |
| 1. Caking Coal |  |  |
| 2. Coal Fines |  |  |
| B. Non-Water Jacket Construction* (Hi-press.) | 8 - 10 | 3.4 |
| C. Large Diameter (assume 18 ft, Low-press.) | 8 - 10 | 7.1 |
| TOTAL ESTIMATED COST (Gasifier Islands Only) |  | 13.0 |

*10 ft diameter gasifier.

Figure 4. Fixed bed gasifier improvement

$13 million. Developmental effort for the fluidized bed gasifier, figure 5, and the entrained bed gasifier, figure 6, is considerable, $103 and $125 million, respectively. In addition to costs for the gasifier proper, miscellaneous support development (figure 7) was estimated at approximately 25 percent of the total gasifier proper development, $70 million. In figure 8, the time and money requirements for cold gas cleanup application and hot gas cleanup development are shown at approximately $5 and $30 million, respectively. In summation of the above, over the next 10 to 15 years, research and development effort to accomplish the proposed goals would place overall costs in the neighborhood of $350 million.

We Americans have great expectations, and in our short history we have generally realized our expectations in a relatively short time. Unfortunately, research and development in the field of coal conversion will require both time and money. We could not forsee the difficult problems concerning energy and the environment, so now we are faced with crash programs. However, coal conversion development is very difficult and will not proceed as quickly as many would like. With coal conversion processes, we are immersed in the real and dirty world, and we must yield processes that perform reliably for 20 to 30 years.

Since our energy demands are here, we should implement the commercially available processes for coal conversion wherever possible and proceed, this time, with a realistic research and development program to let us be prepared for the future. Since we were behind before we started, the crash development approach is necessary; however, for backup we should also concurrently proceed with the conservative approach to insure against complete failure in the event that we are unable to attain the ambitious goals. We feel that onsite generation of low-Btu gas is an attractive coal conversion approach that should be fully investigated to determine its role in supplying our energy needs.

### *Acknowledgment*

We would like to thank the Office of Coal Research, Department of the Interior, for its cooperation in the writing of this paper.

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| | To Commercially Demonstrate | |
|---|---|---|
| | Estimated Time Frame, Years | Estimated Cost In 1974 Dollars $MM |
| A. One Stage Atmospheric Gasifier*, 1 unit | 4 - 6 | 3 |
| 1. High carbon utilization<br>2. Air as oxidizer | | |
| B. Two Stage Atmospheric Gasifier*, 1 unit | 6 - 8 | 5 |
| 1. Hot char recycle<br>2. Air as oxidizer | | |
| C. One Stage Pressure Gasifier (300-500 psig)**, 1 unit | 10 - 12 | 25 |
| 1. Caking coal feed capability<br>2. Coal feed and ash removal capability<br>3. Non-water jacket construction<br>4. Air as oxidizer | | |
| D. Two Stage Pressure Gasifier (300-500 psig)**, 2 units | 12 - 15 | 70 |
| 1. Hot char recycle<br>2. Caking coal feed capability<br>3. Coal feed and ash removal capability<br>4. Non-water jacket construction<br>5. Air as oxidizer | | |
| TOTAL ESTIMATED COST (Gasifier Islands Only) | | 103 |

* 5 ton/hr gasifier for industrial heating application

** Gasifier island to supply thermal output for a 100 MW power plant

Figure 5. Fluid Bed Gasifier Development

| | To Commercially Demonstrate* | |
|---|---|---|
| | Estimated Time Frame, Years | Estimated Cost In 1974 Dollars $MM |
| A. One Stage Atmospheric Gasifier, 1 unit | 4 - 6 | 15 |
| 1. High carbon utilization<br>2. Air as oxidizer | | |
| B. Two Stage Atmospheric Gasifier, 1 unit | 6 - 8 | 20 |
| 1. Hot char recycle<br>2. Air as oxidizer | | |
| C. One Stage Pressure Gasifier, (300-500 psig), 1 unit | 12 - 14 | 25 |
| 1. Coal feed and slag removal capability<br>2. Non-water jacket construction<br>3. Air as oxidizer | | |
| D. Two Stage Pressure Gasifier (300-500 psig), 2 units | 14 - 16 | 65 |
| 1. Hot char recycle<br>2. Coal feed and slag removal capability<br>3. Non-water jacket construction<br>4. Air as oxidizer | | |
| TOTAL ESTIMATED COST (Gasifier Islands Only) | | 125 |

* Gasifier island to supply thermal output for a 100 MW power plant

Figure 6. Entrained Bed Gasifier Development

A. Hot Char Valves

B. Metallurgy

  1. Pressure vessel containment
  2. Corrosion-erosion studies

C. Refractory

  1. Slagging stage application
  2. Selection and installation

D. Pressure Systems

  1. Hot char recycle
  2. Coal feed
  3. Slag removal

E. Surge Volume and Controls for Combined Cycles

F. Gasifier Related Instrumentation and Control Devices

G. Hot Gas Valves and Flanges

H. Kinetic-Thermodynamic Studies

I. System Economic and Operation Studies

J. Environmental Impact

K. Safety Analysis

Estimate cost at 25 percent of gasifier island development, $70 MM.

Figure 7. Miscellaneous support development

| | To Commercially Demonstrate | |
|---|---|---|
| | Estimated Time Frame, Years | Estimated Cost In 1974 Dollars $MM |
| A. Cold Gas Cleanup* | | |
| 1. Sulfur removal ($H_2S$, COS, $CS_2$) systems, 2 units | 4 - 6 | 2.4 |
| 2. Particulate removal systems, 2 units | 4 - 6 | 0.8 |
| 3. Waste water cleanup systems, 2 units | 4 - 6 | 1.6 |
| TOTAL ESTIMATED COST | | 4.8 |
| B. Hot Gas Cleanup - Pressure Systems** | | |
| 1. Hot sulfur removal ($H_2S$, COS, $CS_2$) systems, 2 units | 10 - 14 | 20 |
| 2. Hot particulate removal systems, 2 units | 10 - 14 | 10 |
| TOTAL ESTIMATED COST | | 30 |

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* Cost and time frames included in gasifier island development. Estimated 2 year demonstration on fixed bed units.

** Added cost to gasifier island development for pressure no-tar producing 100 MW electric power output capacity coal gas producers.

Figure 8. Gas cleanup application/development

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# ENVIRONMENTAL ASPECTS OF COAL LIQUEFACTION

P. M. Yavorsky and Sayeed Akhtar*

*Abstract*

*The flow system and all effluents will be described for the SYNTHOIL process, as typical of any coal-to-oil conversion plant that incorporates sulfur removal from the product. Disposal of sulfur, waste off-gas, and waste solid residues will be discussed. The potential hazards of contaminants in product oil and in any leachable residues will be examined to show where chemical data and/or process design alternatives may be needed.*

## INTRODUCTION

The urgency of developing processes for converting coal into clean liquid fuels is widely recognized and research has advanced to the stage that substantial private funds may soon be forthcoming for development. Unfortunately, research on the environmental pollution problems arising from coal liquefaction has received little attention. The significance of this shortcoming can hardly be overemphasized; lack of technology for the control of pollutants will seriously impede the development of coal liquefaction. The siting of even small pilot and first generation plants may present monumental difficulties. It is the purpose of this paper to review the sources of pollution in coal liquefaction, the status of control technology, and the areas of research and development. Research in some of these areas may also be helpful in advancing the science and technology of coal liquefaction itself.

## COAL LIQUEFACTION

The basic steps in a coal liquefaction process are shown in figure 1. Although drawn for the SYNTHOIL process of the U.S. Bureau of Mines, the figure with minor modification can be made to represent other coal liquefaction processes as well. The pollution problems of the pyrolytic processes for making liquid fuel from coal are not a part of this study.

The object of the SYNTHOIL process is a convert coal to a low-sulfur fuel oil. The novel feature of the process is that the liquefaction and hydrodesulfurization are conducted in a turbulent-flow packed-bed reactor (refs. 1,2,3). Slurries of coal in recycle oil and $H_2$ are passed concurrently through a reactor packed with pellets of $Co\text{-}Mo/SiO_2\text{-}Al_2O_3$ catalyst at 450°C and 2,000 to 4,000 psi. The flow of $H_2$ through the reactor is turbulent, which prevents plugging of the reactor, promotes $H_2$ transfer to reaction sites, and facilitates heat removal. Coal is thus liquefied and desulfurized in one step. The product stream from the reactor is cooled and led to a gas disengager to separate the gases from the liquids and unreacted solids. The separated gases, after purification, are recycled to the reactor, and the liquids are centrifuged to remove the unreacted solids. The centrifuged liquid product is a low-sulfur, low-ash fuel oil, a portion of which is recycled to convey more coal to the reactor, while the rest is available as the net product. The solids are pyrolyzed to yield an additional quantity of nonpolluting fuel oil. The total yield of oil is thus about 3 bbl/ton of coal (as received): 2.5 bbl from the centrifuge and 0.5 bbl from the pyrolyzer. The carbonaceous residues from the pyrolyzer and some coal are gasified to product $H_2$ for the process requirement.

## ENVIRONMENTAL IMPACT

The sources of pollution from a coal liquefaction plant and the R & D needs are as follows:

### *Gaseous Effluents*

The off-gas from a coal liquefaction reactor contains a number of impurities which must be scrubbed before recycling the gas. Accurate analytical data on the impurities are not available, but the principal impurities are known to be $CH_4$, $C_2H_6$, $H_2S$, $NH_3$, and $H_2O$. A suitable scrub liquid for $H_2S$ and $NH_3$ is water, and for the gaseous hydrocarbons, an oil. Scrubbing will be conducted at the plant pressure, and the dissolved gases will be recovered from the scrub liquid by a pressure and/or temperature swing. The recovered gases will be processed to separate $NH_3$ as a salable byproduct, while $H_2S$ will be converted to elemental S for ease of storage. The hydrocarbon gases will be

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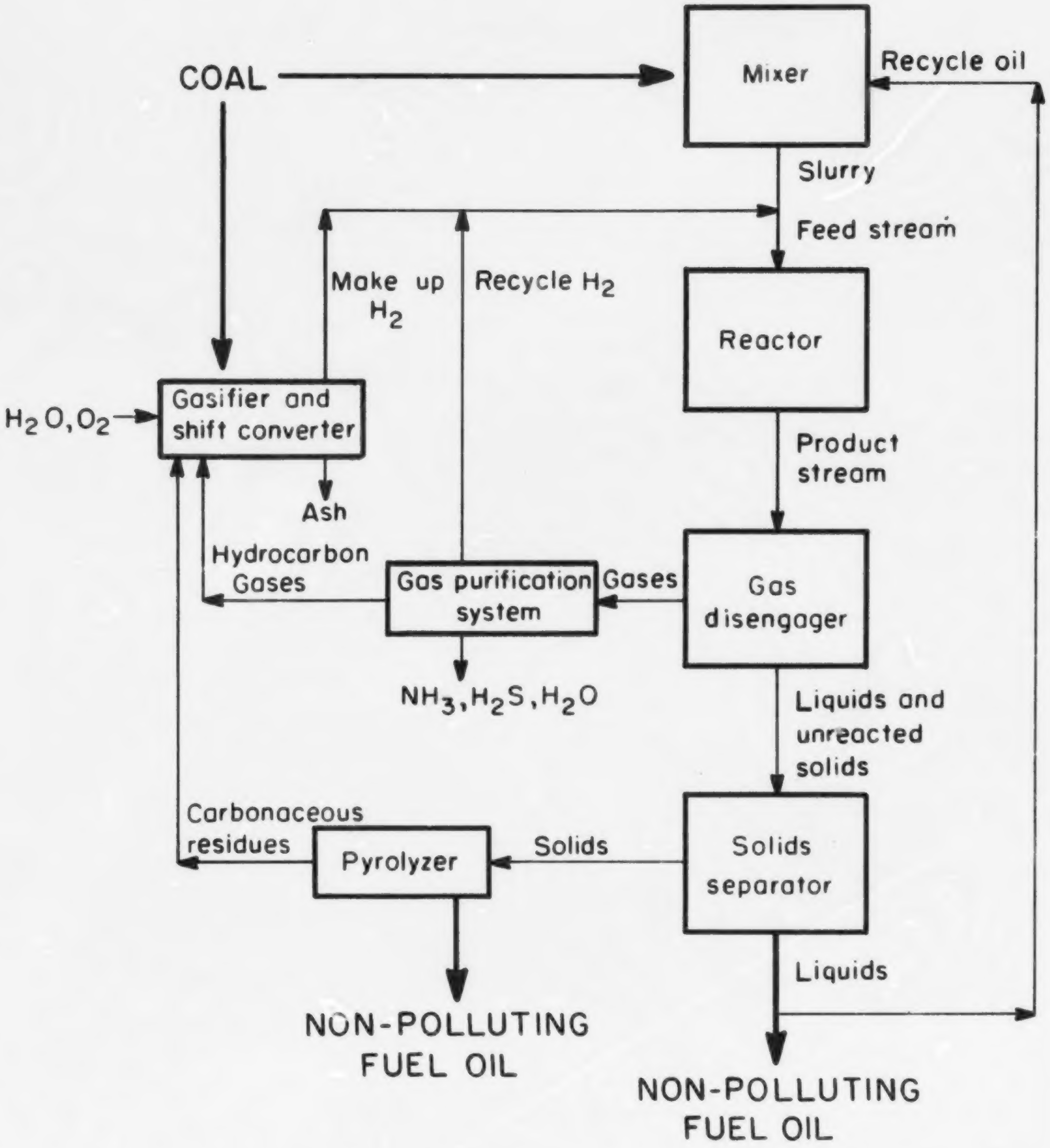
COAL
Mixer
Recycle oil
Slurry
Feed stream
Make up $H_2$
Recycle $H_2$
Reactor
$H_2O, O_2$
Gasifier and shift converter
Ash
Product stream
Hydrocarbon Gases
Gas purification system
Gases
Gas disengager
$NH_3, H_2S, H_2O$
Liquids and unreacted solids
Carbonaceous residues
Pyrolyzer
Solids
Solids separator
Liquids
NON-POLLUTING FUEL OIL
NON-POLLUTING FUEL OIL

Figure 1. Synthoil process

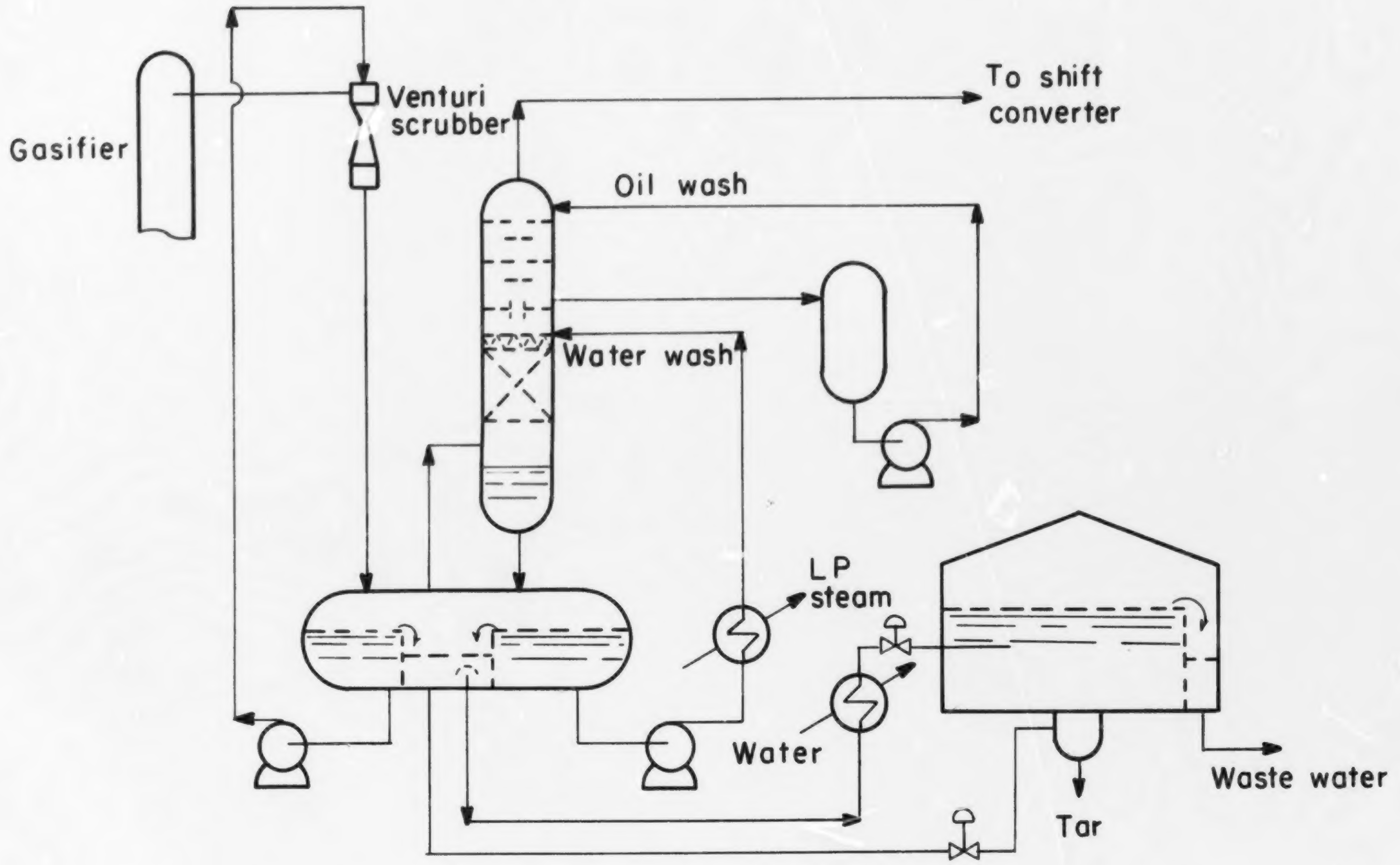
Gasifier
Venturi scrubber
To shift converter
Oil wash
Water wash
LP steam
Water
Waste water
Tar

Figure 2. Raw product gas scrubbing

steam-reformed to generate makeup $H_2$. The technologies for conversion of $H_2S$ to S and for stream-reforming of hydrocarbon gases are commercial, but the rest of the system needs to be developed.

Another source of gaseous effluents will be the impurities in the gasification products of carbonaceous residues and coal. The Bureau is currently conducting an intensive investigation of these impurities and methods of scrubbing them (ref. 4). Figure 2 is a diagram of the scrubbing system being installed on the SYNTHANE pilot plant. The scrubber consists of two sections: a water scrubber to remove the N- and S-containing compounds, and an oil scrubber to remove the hydrocarbon gases and vapors. The system will operate at 1,000 psi. The recovered gases from this source may be combined with those recovered from the recycle gas for processing.

The liquefaction plant will also have gaseous effluents from the mixer, the solids separator (the flash gases), and the pyrolyzer. These will consist mainly of hydrocarbon gases and vapors, $H_2$, $NH_3$, and $H_2S$. They will be added to the gasification products of carbonaceous residues and coal to scrub the impurities and utilize the $H_2$ as makeup gas.

### *Liquid Effluents*

The liquefaction plant will have scrub-water and scrub-oil effluents from the recycle gas purification system and from the makeup $H_2$ generation section. Development of suitable methods for purifying these waste liquids for reuse is essential for both environmental and economic reasons. Analytical data for the waste water from the gasification of different coals have recently been published by the Bureau and work on treatment of the water is in progress (ref. 4). Similar studies of the waste water from recycle gas scrubbing and of the waste scrub oils are necessary to develop purification systems.

As shown in figure 2, there will also be some byproduct tar from the gasifier for makeup $H_2$. Depending upon the type of gasifier and the rank of the coal gasified, the yield of tar is 60 to 80 lb/ton of coal. Also, depending on the coal, the S content of the tar is 1.1 to 2.8 percent (ref. 4). We have found that the tar can be hydrodesulfurized to an environmentally acceptable fuel by the SYNTHOIL process. The properties of a sample of tar from the SYNTHANE gasifier and of the hydrodesulfurized oil from it are given in table 1. The product contained 0.56 percent S, compared with 1.8 percent S in the tar. The yield of oil was 90 percent by weight of the tar, and $H_2$ consumption was 1,088 scf/bbl of oil. The hydrodesulfurization was conducted at the relatively mild conditions of 2,000 psi and 425°C. An oil containing 0.1 to 0.2 percent S can be obtained by hydrodesulfurizing the tar at 4,000 psi and 450°C, but the $H_2$ consumption will be appreciably higher.

Table 1. Hydrodesulfurization of SYNTHANE gasifier tar

Temperature: 425° C
Pressure: 2,000 psi
Catalyst: $Co\text{-}Mo/SiO_2\text{-}Al_2O_3$ (1/8-inch x 1/8-inch pellets)

| | Tar | Hydrodesulfurized oil |
|---|---|---|
| S, wt pct ............. | 1.8 | 0.56 |
| Viscosity, SSF at 77° F | 251 | 15 |
| Specific gravity, 60°/60° F ....... | 1.119 | 1.040 |
| Calorific value, Btu/lb | 14,920 | 16,375 |

Yield: 90 pct by wt
Consumption of $H_2$: 1,088 scf/bbl of oil

### *Solid Wastes*

There will be two types of solid wastes from a coal liquefaction plant: ash and sulfur. Until large new markets for sulfur develop, we are inclined to classify it as a waste. Both ash and sulfur may be disposed of in worked-out coal mines. Alternatively, the ash may be used as a landfill. The question of water-solubles in ash is therefore important and should be studied.

### *Trace Metals in Oil*

A knowledge of the trace metals in coal-derived liquid fuels is important, since the metals will end up in stack gases during combustion. The information is also relevant to problems of fireside corrosion.

An analysis of the metals in a sample of SYNTHOIL is given in table 2. The oil was ashed at 550°C, and the ash was analyzed for silica and metals. The concentrations are reported as ppm in the oil. Silica is present in highest concentration, followed by iron and aluminum.

### *Dust*

Finely divided solids will become airborne during

Table 2. Trace metals in SYNTHOIL

| S in oil, wt percent | 0.22 |
|---|---|
| Ash in oil, wt percent | 0.03 |
| | Concentration in oil, ppm |
| Silica | 135.5 |
| Iron | 67.4 |
| Aluminum | 29.1 |
| Potassium | 5.0 |
| Sodium | 2.9 |
| Calicum | 2.3 |
| Magnesium | 2.2 |
| Molybdenum | <0.2 |
| Cobalt | <0.1 |

(1) transfer of coal from delivery trains to storage hoppers, (2) pulverization of coal, and (3) transfer of ash and S from storage hoppers to disposal trains. The methods of coal dust control, to the extent that they are currently practiced, are based on the use of baghouse filters (widely used in coal-crushing facilities), suction devices (frequently installed between conveyor belts and in closed areas), and water sprays (for suppressing dust from coal piles). Among these, the baghouse filter is the most efficient device; although we are not aware of its application to transfer operations of types (1) and (3) above, we do not consider such applications major problems of development.

### *Carcinogens in Coal Liquefaction Products*

Coal liquefaction products have long been suspected to contain carcinogenic compounds, but no definitive studies are reported. With the list of carcinogens published by OSHA as a guide, their presence in coal liquefaction products should be investigated and appropriate recommendation for occupational safety developed.

## ACKNOWLEDGMENTS

We are thankful to Mr. A. J. Forney and Mr. A. W. Deurbrouck of Pittsburgh Energy Research Center for helpful discussions. To Mr. Forney we are also thankful for permission to use figure 2. The analysis of SYNTHOIL for trace metals was conducted by Dr. Richard C. Diehl of Calgon Corporation. The complete results of our studies with Dr. Diehl will be published separately.

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Disposal of liquefaction plant waste

| Material | Method |
|---|---|
| Sulfur | Claus conversion to elemental form. Surface fill or storage, inert. |
| Off-gass | Reprocess through hydrogen generating gasifier. |
| Solid residue | Land fill or mine fill, like ash. |

## Liquefaction processes differ only in reactor subsystems

1. Synthoil - Direct coal hydrogenation in fixed bed of catalyst.
2. H-coal - Direct coal hydrogenation in ebullated bed of catalyst.
3. SRC - Hydrogen transfer solvent extraction of coal with $H_2$ added to solvent in reactor.
4. Consol-SF - Extraction with separate hydrogenation of solvent and heavy extract.

Common to all are: coal prep., oil handling, residue disposal.

## Anticipated environmental concerns--specific for coal liquefaction plants

| Plant effluent | Potential pollutant | Needed research |
|---|---|---|
| Product oil | Organic carcinogens | A. Analyze for known carcinogens.<br>B. Test oils on animals. |
| Product oil | Metal compounds | A. Analyze for metals.<br>B. Analyze for organometallics. |
| Product oil | Inhalable vapors | Analyze for health hazards. |
| Vapor leaks in slurry prep., residue take-off | As above | As above. |
| Inorganic residue disposal | Leachable inorganic compounds | Analyze for soluble metals, sulfur, halides, etc. |
| Dissolved gases in scrub water | HCN, Se, As, F | Analyze liquor. |

# POTENTIAL BYPRODUCTS FORMED FROM MINOR AND TRACE COMPONENTS IN COAL LIQUEFACTION PROCESSES

**Philip S. Lowell and Klaus Schwitzgebel***

*Abstract*

*Simplified concepts are given for coal liquefaction processes which broadly define processing conditions and objectives. The fate of trace elements in coal liquefaction will be determined by the interaction of coal constituents and process conditions.*

*Minor components such as sulfur, nitrogen, and the trace element selenium were used as examples for identifying the problems associated with determining the fate of trace elements in liquefaction processes. Some research areas for the trace element problem can be specified at this time. These specific areas will lead to treatment process problem definitions. The research needs for actual development of treatment processes cannot be detailed at this time.*

## INTRODUCTION

The need for an environmentally acceptable form of coal has been amply justified. The major objections to the direct use of coal as a fuel in the United States today are the particulate and sulfur oxide emissions.

There are many processes for converting coal to liquid or gaseous products. Each process has various features that may be more or less advantageous in any given situation.

Yavorsky (ref. 1) listed the following distinguishing advantages for liquefaction processes:

(1) The carbon/hydrogen ratio must only be increased slightly (0.9 to perhaps 1.1);
(2) Liquid products are easily storable;
(3) Liquid products can serve as chemical feed stocks as well as fuels.

The developers of liquefaction processes are dedicated to optimizing the principle process goal; the production of the liquid products. For these products to be saleable, the sulfur and nitrogen contents must be below certain levels. Therefore, considerable effort has been directed toward sulfur and nitrogen removal.

The fate of the various trace elements in coal is not in the "mainstream" of process development. This is probably rightfully so in the initial stages. But the time has come when coal processing plants must solve these problems in order to be environmentally acceptable themselves. The chemical form and the streams in which minor and trace elements appear will be determined by the interaction of their chemical combination in the coal with the processing conditions. The problem faced now is to have treatment processes available when they are needed.

To define the research and development needs for the environmental aspects of trace elements in coal liquefaction processes, we should perhaps look at a liquefaction plant as we envision the way it will be built and work backwards to where we are today. When a vast coal processing complex (ref. 2) is finally built, we would like to be able to know in which outgoing streams and in what form all of the incoming trace elements will leave the process. Some elements may leave in a harmless form with the solid waste ash (e.g., phosphorus as phosphates). Others may leave with the product, in concentrations low enough to be harmless and/or in nontoxic chemical forms. Others may be converted to highly toxic or odorous forms and require removal processes. With this information, the design engineer would like to get out his design manual, with its tabulations of physical and chemical data, and design the required pollutant removal processes.

Development of trace element treatment processes cannot logically be started or the scope defined until the stream conditions and chemical form are established. The development of sampling techniques and analytical chemistry methods is a necessary step. The sampling and analysis methods must be used on pilot plants to determine these data.

We are in a "chicken and the egg" situation. We must know in what streams, in what concentration, and in what chemical form the trace elements will appear so that we can develop sampling techniques and analytical methods to measure them quantitatively. The starting point then is a prediction of the course of phase separations and chemical reactions for the trace elements. Several iterations will be required before success is finally reached.

Another aid in defining research needs is to consider the few elements in coal for which a level of understanding does exist and then to extrapolate this data to the other unknown elements. For instance, the amount of information on sulfur in coal is considerable (although the difficulty of the problem is also great). The types of sulfur bonds have been classified (refs. 3,4,5) and pure compound catalytic and noncatalytic desulfurization tests (ref. 6) have been made.

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On the other hand, the knowledge of nitrogen structures in coal is very sparse (ref. 3). This is surprising in view of the amount of nitrogen present. Model compound reactions are only in rudimentary stages.

The level of knowledge of most trace elements in coal is very low. The research needs for liquefaction processes in this paper will be identified in the following manner:

(1) We will trace what is known about sulfur and nitrogen with respect to pollution control.
(2) We arbitrarily choose selenium as an example of a trace element and try to determine its fate. The problems identified in this attempt will aid in defining the research needs.
(3) We will attempt to generalize from the above specific examples.

## SIMPLIFIED PROCESS OBJECTIVES

The primary objectives of coal liquefaction plants are to produce a product that is (or has):

(1) a liquid (plus some gases);
(2) a low ash content product;
(3) a low sulfur content;
(4) other (e.g., end use as fuel, chemical feed stock, etc.).

Coal may be classified arbitrarily as consisting of organic and inorganic fractions. Modern coal liquefaction processes slurry the pulverized coal in a hydrogen donor solvent (usually recycle solvent derived from the process itself) and treat the slurry with hydrogen gas, possibly in the presence of a catalyst. Solids consisting of inorganic material from the coal and the undissolved heaviest organic portion of the coal are removed by physical means such as filtration or centrifugation. The liquid portion from this physical separation is the product and may be used as fuel or processed as a synthetic crude oil. The solid from this physical separation contains significant heating value and in many process designs will be burned to provide process heat (ref. 7) or subjected to a gasification process to provide process hydrogen (ref. 2). These solids can be expected to contain not only the trace elements from the inorganic portion of the coal but a large fraction of many of the minor components and trace elements in the organic part of the coal. The sulfur content of this solid is very high (typically 6 percent), so sulfur recovery processes are envisioned. Determination and control of the fate of trace elements in the units of the coal refinery complex processing this material will provide one of the major challenges for the trace element program.

To illustrate the liquefaction process, consider the "demonstration" coal molecule of figure I. In order to convert this large, solid molecule to a liquid, the "bridges" must be broken. The aromatic clusters will be liquids or low-melting solids.

Sulfur and nitrogen are each represented with two different bond types. As will be shown later, the ability of the process to remove these minor components depends on the relative amounts of bond type.

## SIMPLIFIED PROCESS CONDITIONS

The liquefaction processes operate through the mechanism of breaking the connecting bridges or linkages between aromatic clusters. These bridges can be aliphatic or they may contain oxygen, sulfur, or other elements. Attacking these groups in bridges is desirable.

Hydrogenation of olefinic and aromatic carbon, or nonlinking oxygen is undesirable since it consumes hydrogen but does not contribute to liquefaction. Breaking of carbon bonds that result in coking is also undesirable.

Thermodynamically, all of the above reactions are feasible at 300°C. Kinetically they do not proceed rapidly enough to be feasible (ref. 20). Aromatic structures become thermodynamically stable at higher temperature with respect to hydrogenation to form saturated compounds. The free energy change for benzene hydrogenation to cyclohexane is -5.2 Kcal at 227°C and +13.6 Kcal at 427°C. Processing conditions are normally set as hot as possible without excessive rupturing of carbon-carbon bonds to produce coke. This has been found experimentally to be in the range of 450° to 460°C.

Catalysts can play a key role in several areas. One is in breaking the desired bridges without excessive hydrogenation of aromatics and unsaturated compounds. Another is release of undesirable elements in the ring structures, e.g., sulfur. Obviously the type of catalyst used will influence the physical and chemical properties of the product as well as the hydrogen consumption required to attain liquefaction. Present catalysts are kinetically active at about 370°C and above. Hence, the normal temperature range of liquefaction processes are 400° to 450°C.

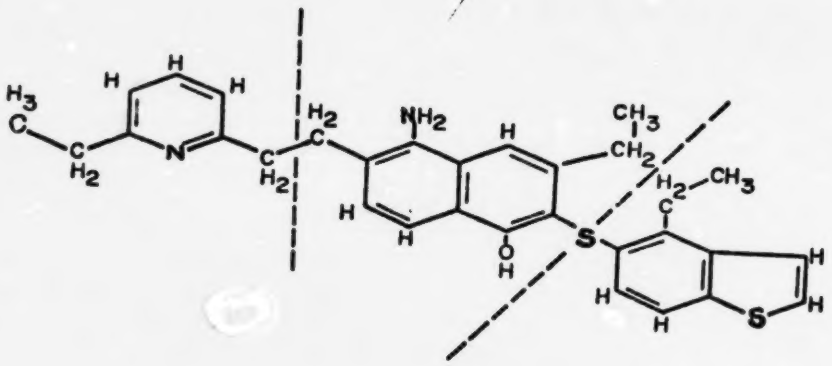


Figure 1. Coal molecule

Table 1. Typical processing conditions

| Process | Temperature °C (°F) | Hydrogen Pressure Atmospheres (PSI) | Catalyst |
|---|---|---|---|
| Solvent Refined Coal (SRC) | 440 | 70 | None |
| Pittsburg and Midway | (825) | (1000) | |
| H-Coal | 455 | 170 | Cobalt Molybdate |
| Hydrocarbon Research, Inc. | (850) | (2500) | |
| Synthoil | 450 | 270 | Cobalt Molybdate on activated $SiO_2$-$Al_2O_3$ |
| Bureau of Mines | (840) | (4000) | |

High hydrogen partial pressure is of value in increasing kinetics as well as in giving a better thermodynamic potential. As will be seen later, this is necessary in some cases.

Process conditions for three important liquefaction processes are summarized in table 1.

## PREDICTIONS OF REACTIONS

Experimental data and thermodynamic calculations are presented for sulfur, nitrogen, and selenium as examples of what may be expected.

### *Sulfur*

Sulfur exists in several forms in coal. Two bond types, a model compound having this type bond, and the Gibbs free energy for the model compound reacting to give $H_2S$ are tabulated in table 2. The free energies were estimated by the van Krevelen and Chermin method (refs. 8,9). The reaction mechanism proceeds through sulfur removal first. Saturation of resulting olefins follows this step (ref. 10).

Experimental data indicate that almost all of the sulfide sulfur will be converted to $H_2S$ at 400°C in a hydrogen atmosphere. At 400°C and 100 atm hydrogen pressure, very little thiophene sulfur is removed unless a catalyst is used. Workers at the Bureau of Mines have reported excellent sulfur removal from a distillate fuel oil product using a fixed bed of cobalt molybdate catalyst (refs. 1,11,12). Johnson et al. (ref. 13) report overall desulfurization of 90 percent using an ebullated bed of cobalt molybdate catalyst in the H-coal process.

In laboratory studies of model compounds, the sulfur in thiophene and benzothiophene can be easily removed to 95+ percent with a catalyst (ref. 6). The addition of a methyl group next to the sulfur on the thiophene ring reduces sulfur removal to a percentage in the mideighties under the same conditions. Apparently the decrease in catalyst efficiency is a steric hindrance problem because the free energy changes for the two compounds are almost the same (see table 2). Dibenzothiophene is much less reactive than the less highly substituted thiophenes (refs. 14,15).

Sulfur appears to follow somewhat predictable rules. The sulfides have the greatest free energy and are most subject to attack. The free energy change of the thiophenes are all of the same order of magnitude. The aromatic sulfur structures are fairly stable except under catalytic attack. Steric factors influence the rate and type of desulfurization.

### *Nitrogen*

Considerably less seems to be known about nitrogen in coals. Nitrogen is more difficult to remove by hydrogenation than sulfur. Possible structure reactions to give ammonia, and free energy changes for the reactions are given in table 3.

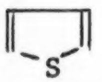
It is of interest to note that the "ease of removal" is in the same order as the free energy changes at 400°C.

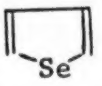
| | $\Delta G^{400°C}$, Kcal |
|---|---|
| sulfide (S) | -30.3 |
| amine (N) | -20.7 |
| ring (S) | ∿ 0.0 |
| ring (N) | + 7.9 |

Thermodynamics will be of help in several ways. First, it can be used to define temperature and pressure areas of interest. Second, it can be used to predict compounds for which one should look. Third, it gives an indication of where catalyst development research would be of value.

### *Selenium*

Selenium is one of the trace components of interest because of its toxicity and malodorous compounds. Chemically, selenium is very similar to sulfur. Selenium compounds analogous to sulfur are known to exist.





| | |
|---|---|
| $H_2S$ | $H_2Se$ |
| $CS_2$ | $CSe_2$ |
| COS | COSe |
| $SO_2$ | $SeO_2$ |
| $SO_4^{=}$ | $SeO_4^{=}$ |

## TABLE 2. SULFUR FORMS AND REACTIONS

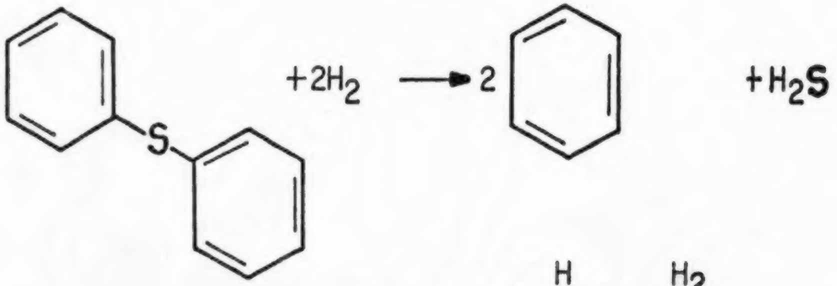
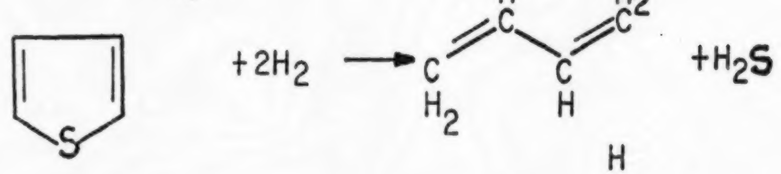
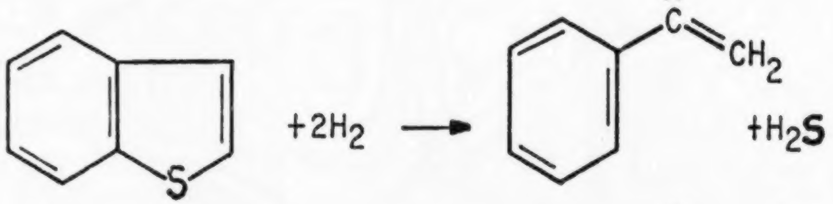
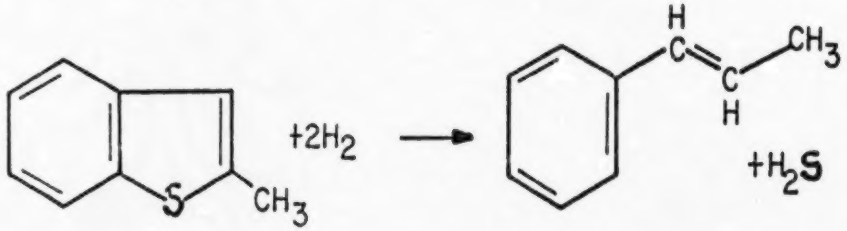
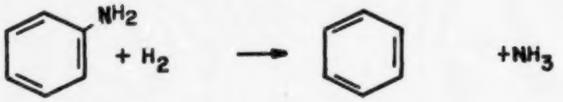
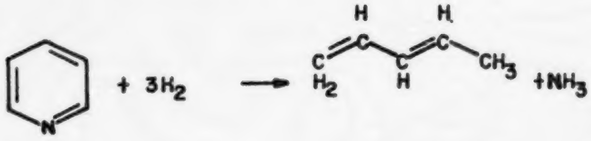
| TYPE OF BOND | MODEL COMPOUND REACTIONS | $\Delta G_R$, K Cal<br>25°C | <br>400°C |
|---|---|---|---|
| SULFIDE |  | --25.8 | -30.3 |
| CONJUGATED RINGS<br>(THIOPHENES) |  | -1.7 | +3.8 |
|  |  | -7.3 | -1.2 |
|  |  | -7.5 | -2.0 |

Table 3. Nitrogen forms and reactions

| TYPE OF BOND | MODEL COMPOUND REACTION | $\Delta G_R$, K Cal<br>25°C | <br>400°C |
|---|---|---|---|
| AMINE |  | −18.1 | −20.7 |
| CONJUGATED |  | −8.4 | +7.9 |

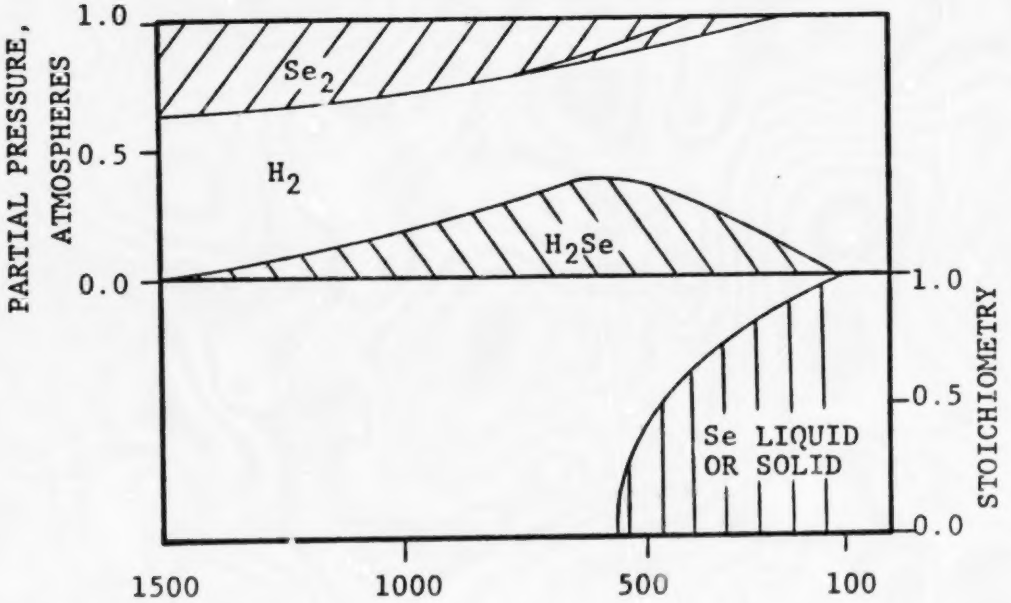


Figure 2. Hydrogen selenide decomposition

The chemical form of selenium in coal is basically unknown. Attari (ref. 16) has reported that selenium leaves the solid phase as gasification proceeds during the HyGas process.

The strengths of the C-Se (ref. 17) and H-Se bonds are less than the corresponding sulfur bonds. DeBerry (ref. 18) has estimated that the free energy of reaction of selenium-carbon compounds to give $H_2Se$ will be less negative by about 5 Kcal than the corresponding sulfur compounds to give $H_2S$. This indicates that the difficulty of selenium removal as $H_2Se$ should be estimated to be between N and S compounds. This assumes that selenium will exist in the same form as sulfur.

Hydrogen selenide is known to be unstable at room temperature. It decomposes to hydrogen and metallic selenium. The vapor pressure of selenium metal is about 3 mm Hg at 400°C. The equilibrium between reactions is reported by Flögel (ref. 19). Flögel's results at 1 atm are summarized in figure 2.

At the high temperatures characteristics of gasifiers, only gaseous $H_2$ and $Se_2$ would exist. We have not investigated compounds such as $CSe_2$, etc. At coal liquefaction process temperatures, $H_2Se$ will be the predominant species. Cooling the mixture further results in decomposition to $H_2$ and liquid or solid selenium.

One would expect some of the selenium to be in the inorganic fraction as selenides, selenites, or selenates. The organically bound selenium will probably leave the process in the liquid product as well as $H_2Se$ in the gas phase.

The gas phase sampling and analysis will be a difficult operation at best. The $H_2Se$ can decompose to metallic selenium (which can have a significant vapor pressure). Should it not decompose, it must be remembered that hydrogen selenide is water soluble (about 2,000 times stronger acid than $H_2S$) and is a moderate reducing agent (ref. 17). The actual chemical analysis technique may well be simple compared to getting a truly meaningful sample in proper condition in a proper container to place before an instrument.

## RESEARCH AND DEVELOPMENT NEEDS

Research programs in energy must be designed to fulfill two important requirements. The information developed must be useful to those who are providing overall management and leadership to the development of energy sources; and the information must be placed in the hands of those who must ultimately implement or commercialize new energy technology.

Both goals can be achieved by a contract research and development program administered by the Federal Government. Leadership and management can be supplied by the appropriate Federal agency, and the participation of industry in the development of new technology will insure the implementation of that technology in the private sector.

In addition to defining research areas it is almost as important to define the type of organization most suited to do the work. This will be indicated also.

### *Organic/Inorganic Portions*

For every trace element the relative amounts that are contained in the organic and inorganic fractions should be determined. Those trace elements that are removable to an acceptable level by pretreatment processes will require only minor additional research. For process or site conditions opting for no pretreatment, further work will be required. This should be done by an industrial organization.

### *Development and Testing of Sampling and Analytical Methods*

An iterative procedure of estimating the form and concentration of trace elements, developing sampling and analysis procedures, testing these procedures, attempting to account for the material balance, revising the estimates, etc., must be done for each process and each trace element. While the material balance should be quantitative, the compound identification will only be tentative.

The bulk of this work should be industrial, but universities have a contribution to make in basic reactions and phenomena upon which analytical methods can be based.

### *Structure Identification*

After the broad outlines of the material balance have been fixed, the actual compounds involved must be identified. Trace element form in the product streams must be identified so that treatment processes may be developed as soon as possible. The trace element structures in coal are of value too.

Bothe industry and universities should participate in the general structure analysis, e.g., cyclic structures, reactive groups, etc. Universities should have prime responsibility for model compound synthesis and reactions. Industry should be concerned with the treatment process aspects.

### *Development of Catalysts*

Historically, catalysts prepared from various combinations of Group VIII and Group VI-B oxides and sulfides have been used as desulfurization catalysts in the petroleum industry. Their effectiveness in cleaning liquid fuels from coal liquefaction processes must be investigated and new catalyst development initiated for selective desulfurization of heavy, coal-derived liquids. This may well require the development of radically new catalytic systems.

Catalysts for nitrogen and trace element removal should also be investigated. For the most part, this will be virgin territory.

This task should be done by both industry and universities. It is an excellent example of an area in which joint industry/university efforts could be particularly advantageous.

### *Physical and Chemical Properties*

Thermodynamic properties and physical data must be gathered and generated for the basis of accurate process designs. The bulk of this research should be done by universities, although industry may have to generate significant portions because of time constraints.

### *Treatment Process Development*

This important task can be identified but, due to lack of knowledge, cannot be defined at this point because too little is known. Because of time considerations, the initial effort will be by industry. Universities will contribute to second generation processes through the work done in "Structure Identification" and "Physical and Chemical Properties."

## ACKNOWLEDGMENTS

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# OIL SHALE AND ITS POTENTIAL UTILIZATION

G. U. Dinneen*

*Abstract*

*The large deposits of oil shale in the Green River Formation in Colorado, Utah, and Wyoming offer a potential source of significant quantities of liquid fuels. This paper discusses the location and potential of the resource; the present state of technology for producing shale oil by both aboveground and in situ processes; the characteristics of shale oil; environmental aspects of oil-shale utilization, and recent developments, particularly results of the Department of the Interior's Prototype Oil Shale Leasing Program, that suggest inauguration of commercial oil-shale processing.*

## INTRODUCTION

Oil shales of the Green River Formation in Colorado, Utah, and Wyoming comprise one of the largest deposits of hydrocarbons in the world and are a potential source of significant quantities of liquid fuels. They are sedimentary rocks containing solid organic material that can be decomposed by heat to yield an oil which can be refined to yield the products normally obtained from petroleum.

Various attempts have been made since before World War I to develop commercial processes for utilizing Green River oil shale. Most of these have involved mining the shale and processing it in aboveground equipment; but recently, in situ processing has received considerable attention. The first approach is reasonably well developed and will probably be used where the shale can be readily mined. The second approach requires more research, but it may have an economic advantage; it may be applicable to deposits of various grades and thicknesses that do not readily lend themselves to mining, and it avoids the problem of disposing of large amounts of spent shale. Both approaches have potential environmental effects that must be taken into consideration.

Although there is no present oil-shale industry in the United States, the current status of oil-shale technology and the country's need for new energy sources suggest that the start of such an industry may be imminent. This paper describes the availability and potential of oil shale, the current technology for producing and utilizing shale oil, the potential environmental effects of oil-shale utilization, and recent developments suggesting commercial development.

## LOCATION AND POTENTIAL OF OIL SHALE

Oil shales are widely distributed throughout the United States, but the largest and richest deposit is in the Green River Formation. Hence, most efforts to utilize oil shale have been on material from this formation, and it will thus be the one considered in this paper. The formation, whose location is shown in figure 1, covers an area of about 17,000 square miles in four principal basins: The Piceance Creek Basin of Colorado, the Uinta Basin of Utah, and the Washakie and Green River Basins of Wyoming. In this formation, oil-shale intervals that are at least 10 feet thick and that yield at least 25 gallons of oil per ton have a potential oil yield in place of 600 billion barrels as shown in table 1 (ref. 1). If thinner shale as lean as 10 gallons per ton were used, the potential oil yield would increase to some 2 trillion barrels.

Although the Piceance Creek Basin, as shown in figure 1, contains only a small part (about 10 percent) of the area covered by the Green River Formation, it contains about 80 percent of the richer oil shale (table 1). The oil shale crops out in cliffs (fig. 2) along the southern edge of this basin. The Mahogany zone, which is the best known interval of the Green River Formation, is about 75 feet thick in the lower part of the cliff just above the talus slope. The rich oil shale thickens toward the center of the Piceance Creek Basin so that in some places continuous sections of oil shale averaging more than 25 gallons of oil per ton are hundreds of feet thick (ref. 2). However, these are generally under several hundred feet of overburden and therefore may not be as readily mined as the outcrops which have received most attention in the past. In Utah and Wyoming, the sections of rich shale are not as thick as those in Colorado, and in Wyoming they are often interspersed with alternating beds of lean shale. Hence, somewhat different recovery techniques may be required for these shales than for those in the Piceance Creek Basin.

A major problem in utilization of oil shale is the

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necessity of handling large amounts of rock than contain only moderate amounts of organic material, as indicated in table 2. Fortunately, the organic matter is fairly high in hydrogen so that about

A major problem in utilization of oil shale is the necessity of handling large amounts of rock than contain only moderate amounts of organic material, as indicated in table 2. Fortunately, the organic matter is fairly high in hydrogen so that about two-thirds of it can be converted to oil by heating. Unfortunately, it has a rather high content of nitrogen which appears in the oil and must be removed by special techniques before the oil can be considered a desirable refinery feedstock.

## TECHNOLOGY

Shale oil may be recovered from the Green River Formation by two general approaches. The first includes mining, crushing, and aboveground retorting; this approach has been used in various parts of the world for over 100 years for the commercial production of shale oil and has been studied in this country for many years. It will probably be used for

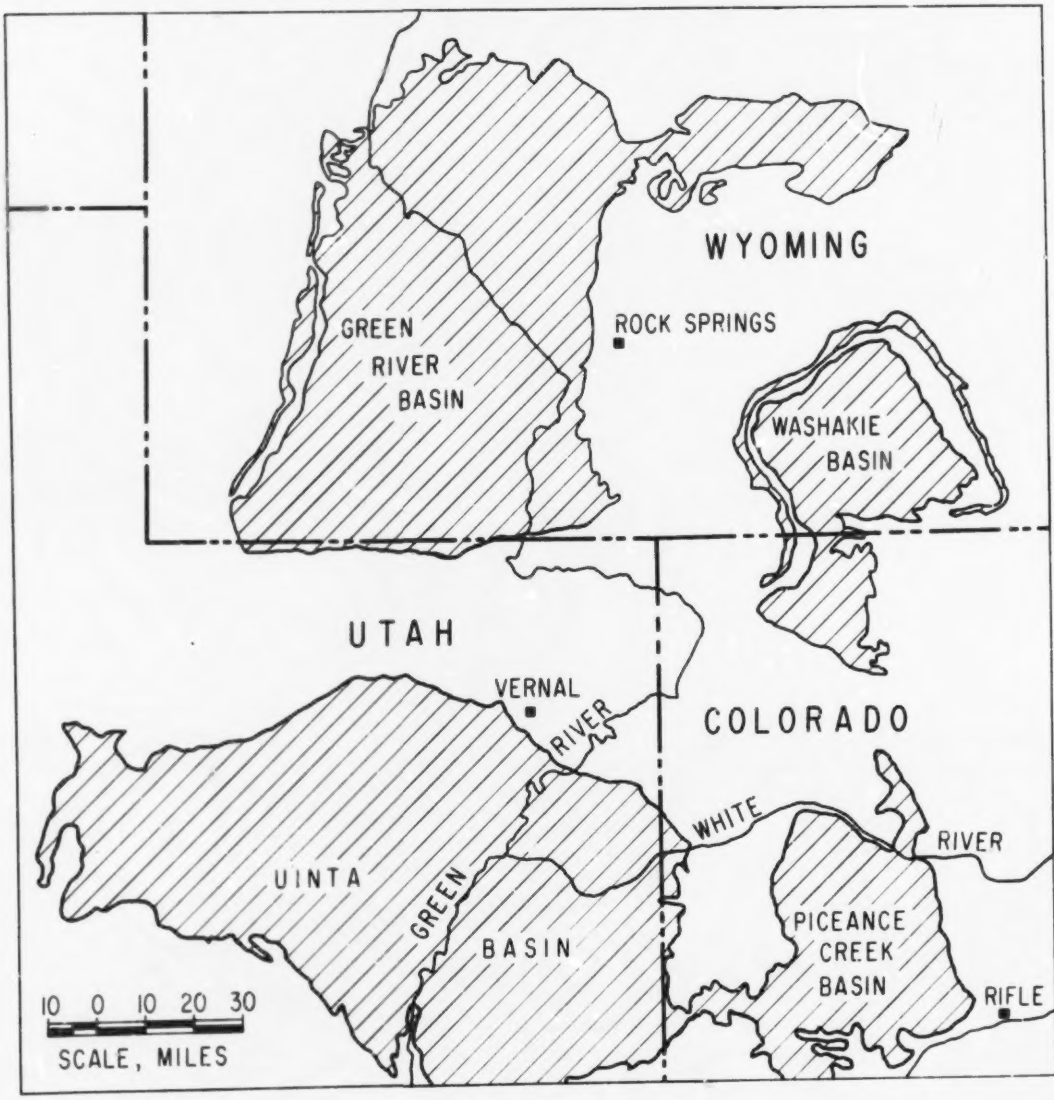
WYOMING
GREEN RIVER BASIN
ROCK SPRINGS
WASHAKIE BASIN
UTAH
VERNAL
RIVER
COLORADO
WHITE
RIVER
UINTA
GREEN
BASIN
PICEANCE CREEK BASIN
RIFLE
10 0 10 20 30
SCALE, MILES

Figure 1. Three-State map.

Table 1. Potential shale oil in known deposits of the Green River Formation

| | Billions of barrels of oil in place | | | |
|---|---|---|---|---|
| | Colorado | Utah | Wyoming | Total |
| Intervals 10 or more feet thick averaging 25 or more gallons of oil per ton | 480 | 90 | 30 | 600 |
| Intervals 10 or more feet thick averaging 10 to 25 gallons of oil per ton | 800 | 230 | 400 | 1,430 |
| Intervals 10 or more feet thick averaging 10 or more gallons of oil per ton | 1,280 | 320 | 430 | 2,030 |

Table 2. Composition of mahogany zone shale of Colorado and Utah

| | Weight percent |
|---|---|
| Mineral matter: | |
| Content of raw shale | 86.2 |
| Estimated mineral constituents: | |
| Carbonates, principally dolomite | 50 |
| Feldspars | 19 |
| Illite | 15 |
| Quartz | 10 |
| Analcite and others | 5 |
| Pyrite | 1 |
| Total | 100 |
| Organic matter: | |
| Content of raw shale | 13.8 |
| Ultimate composition: | |
| Carbon | 80.5 |
| Hydrogen | 10.3 |
| Nitrogen | 2.4 |
| Sulfur | 1.0 |
| Oxygen | 5.8 |
| Total | 100 |

initial development of the Green River Formation. The second is in situ processing, which has received serious consideration only in the last few years, but which has potential economic and other advantages that make efforts to develop a feasible method worthwhile. It might be used where the shale is deeply buried, where it occurs in relatively thin intervals, where it consists of alternating intervals of rich and lean shale, or where various other circumstances exist so that mining cannot be readily applied. In addition, the technique has the advantage of leaving the mineral residues in place, thus eliminating the disposal problem associated with the aboveground approach. However, it may introduce environmental effects of its own, such as adding soluble materials to groundwaters.

### *Mining and Aboveground Processing*

Many attempts have been made to mine and retort Green River oil shale. A simplified schematic representation of this approach is shown in figure 3. So far, all attempts have been made on a pilot-plant scale, and no prototype commercial unit has been operated. Five of the more extensive investigations that have been conducted or are being conducted are those by (1) the Bureau of Mines, (2) a group of six oil companies utilizing Bureau of Mines facilities, (3) the Paraho Development Corporation, (4) Union Oil Company of California, and (5) the Colony Development Operation.

Investigations of mining and retorting oil shale and of refining shale oil were conducted by the Bureau of Mines near Rifle, Colorado, from 1944 to 1956. A demonstration mine was opened in a 73-foot mineable section of the Mahogany zone, and it was shown with fair assurance that low mining costs and

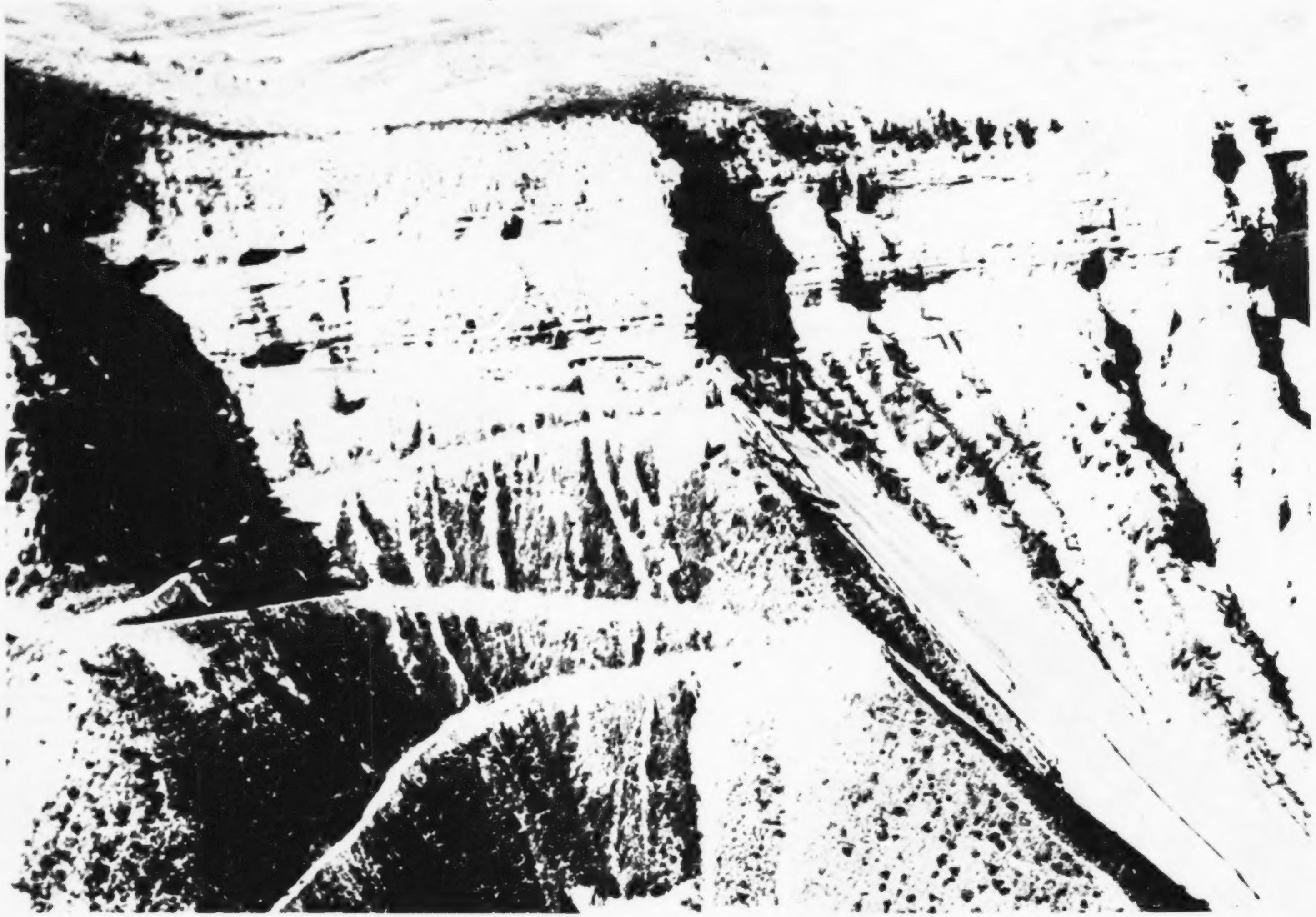

Figure 2. View of cliff.

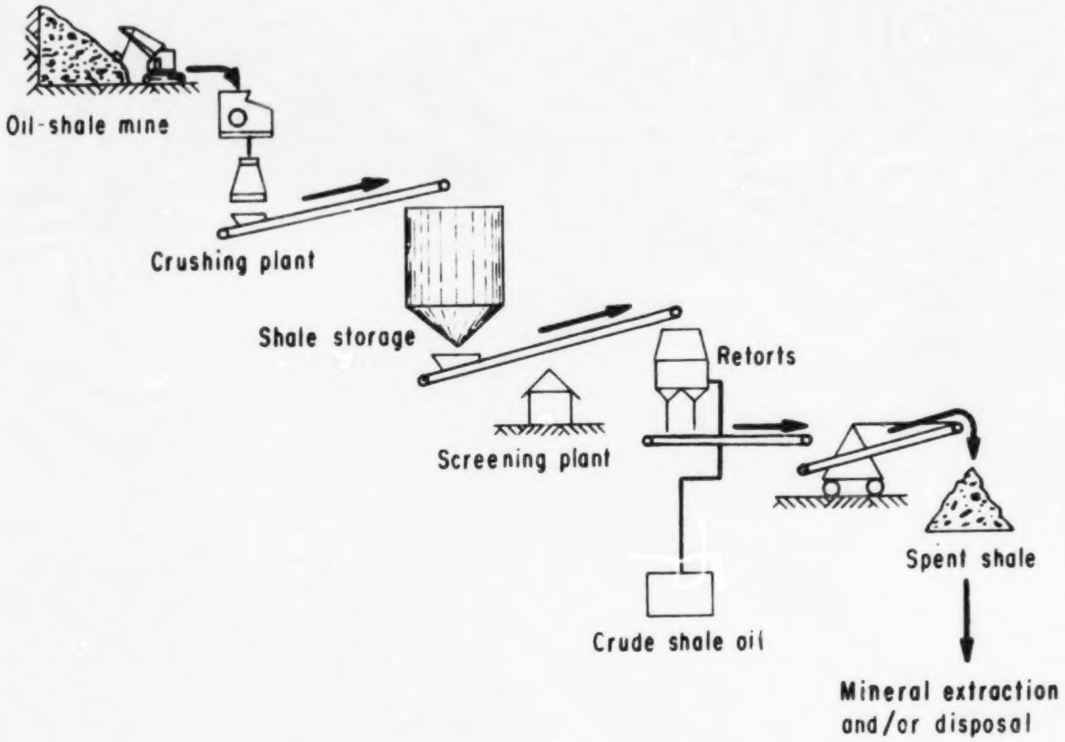

Figure 3. Schematic of oil shale surface processing.

high recovery in a room-and-pillar operation were possible (ref. 3). Refining research provided assurance that petroleum refining technology would be adaptable to shale oil.

During the Bureau's program, retorting research led to development of the gas combustion retort (ref. 4), which was considered the most promising of the retorting methods investigated. This retort, which is shown schematically in figure 4, is a vertical, refractory-lined vessel through which crushed shale moves downward by gravity. Recycled gases enter the bottom of the retort and are heated by the hot retorted shale as they pass upward through the vessel. Air is injected into the retort at a point approximately one-third of the way up from the bottom and is mixed with the rising hot recycled gases. Combustion of the gases and some residual carbonaceous material from the spent shale heats the raw shale immediately above the combustion zone to retorting temperature. Oil vapors and gases are cooled by the incoming shale and leave the top of the retort as a mist. The manner in which retorting, combustion, heat exchange, and product recovery are carried out gives high retorting and thermal efficiencies. The process does not require cooling water, an important feature since the shale deposits occur in semiarid regions. The development program utilized pilot plants having capacities of 6, 25, and 150 tons per day, but was terminated before operability of the largest of these had been completely demonstrated. However, the process appeared to offer the possiblity of large-scale operation.

The gas combustion retorting system was developed further during the period 1964 to 1968 when the Colorado School of Mines Research Foundation leased the Bureau of Mines Rifle facilities and operated them under a research contract with six oil companies: Mobil (which acted as Project Manager), Humble, Phillips, Sinclair, Pan American, and Continental. The research was conducted in two stages, each of which lasted approximately 2 years. The first stage was devoted primarily to investigating the gas combustion retorting process itself in the two

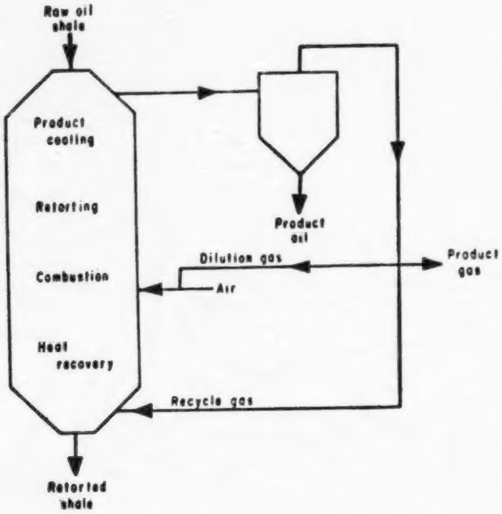

Figure 4. Gas combustion retort.

smaller pilot plants that had been constructed by the Bureau (ref. 5). The second stage included research on both mining and retorting. The mining involved development of a room-and-pillar method similar to that of the Bureau of Mines except that somewhat smaller pillars were used. During this stage it was demonstrated that the largest gas combustion pilot plant could be operated using feed rates of 500 pounds per hour per square foot of cross-sectional bed area, about double the rate previously achieved by the Bureau of Mines, while maintaining oil yields in excess of 85 percent of Fischer assay. Although the results indicated a significant advance in development of the process, some operating problems were not fully resolved (ref. 6).

A modified version of the gas combustion retorting system, designated as to the Paraho retort and successfully applied to calcining limestone, is presently being tested on oil shale in a program supported by 17 companies (ref. 7). Major modifications to the process involve the charging and discharging mechanisms for the retort and the gas injection and process control systems. The test program, which started late in 1973, is scheduled to last 30 months and will cost $7.5 million. Two retorts, a pilot plant 2½ feet in diameter and a semiworks plant 8½ feet in diameter, will be constructed at the Bureau of Mines Rifle facilities, which have been leased by the Paraho group for their test program.

A retorting system developed by Union Oil Company of California also consists of a vertical, refractory-lined vessel. However, it operates on a downward gas flow principle and the shale is moved upward by a charging mechanism ususally referred to as a rock pump. Heat is supplied by combustion of the carbonaceous residue remaining on the retorted shale and is transferred to the oil shale, as in the gas combustion retort, by direct gas-to-solids exchange. The oil is condensed on the cool incoming shale and flows over it to an outlet in the bottom of the retort. This process also has the advantage of not requiring cooling water. The system was tested from 1956 to 1958 on a demonstration scale of about 1,000 tons per day. It was subsequently announced that operation of the plant had yielded enough information so that the process could be commercialized whenever energy demand and economic conditions warranted (ref. 8). Several recent announcements in the press have indicated that Union may start construction of a commercial plant relatively soon, but the announcements did not give details of the processing scheme that will be used.

The process that appears to be nearest to commercial utilization is the TOSCO II system, which is based on a rotary kiln utilizing ceramic pellets heated in external equipment to accomplish retorting. Shale feed which is ground to less than ½ inch in size, is preheated by flue gases from the pellet-heating furnace and introduced into the kiln with pellets heated to 1,200°F. In the kiln, it is brought to a retorting temperature of 900°F by heat exchange with the pellets. Passage of the kiln discharge over a trommel screen permits recovery of the pellets from the fine shale for reheating and recycling. Oil vapors are recovered, and the spent shale is routed to disposal.

The TOSCO II process together with a room-and-pillar mining method has been under investigation for several years in a semiworks plant having a capacity of about 1,000 tons per day. The Colony Development Operation—originally composed of the Standard Oil Company of Ohio, the Oil Shale Corporation, and Cleveland-Cliffs Iron Company, but subsequently including Atlantic Richfield Oil Company as Project Manager—conducted this investigation, which terminated in April 1972. During the investigation, a considerable research effort was also expended on environmental aspects of oil-shale operations, particularly in regard to stabilizing and vegetating spent-shale deposits. After the pilot plant

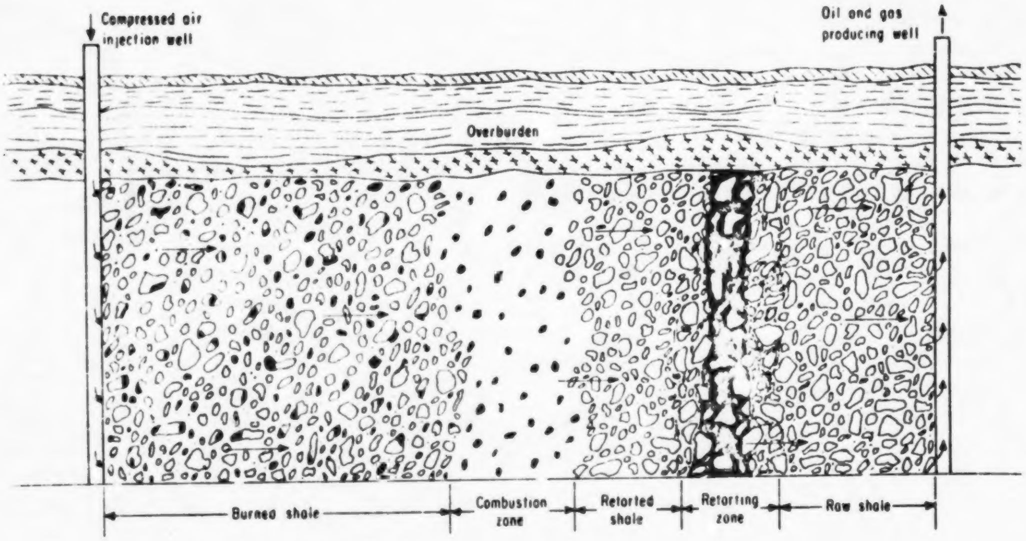

Figure 5. Schematic diagram of an *in situ* oil shale retorting process.

was shut down, results were evaluated and design of a commercial plant having a capacity of about 50,000 barrels of oil per day was started. The engineering design for this plant will be completed later this year, and it has been announced that plant construction could start probably in October if other phases of the project, such as obtaining a permit for a pipeline from the shale plant location to the Four Corners area, can be completed by that time (ref. 7).

*In Situ Retorting*

The process of retorting oil shale in place has been receiving increased attention in recent years because it may have a number of advantages: it may be more economical than the traditional approach; it may be applicable to deposits of various thicknesses, grades, and amounts of overburden that are not readily amenable to mining; and it eliminates the necessity of disposing of large quantities of spent shale. However, it may introduce other environmental problems such as the possibility of groundwater leaching the soluble retorting products left underground. In spite of its potential advantages and the recent interest in it, only a relatively small amount of research has been done on in situ processing; consequently, technology is generally in the early stages of development.

In situ retorting might be accomplished by passing gases and liquids either horizontally or vertically through fractured shale. The horizontal approach is illustrated schematically in figure 5. One application of this approach consists of drilling a predetermined pattern of wells into the oil-shale formation, creating permeability among the wells if naturally occurring permeability is low, igniting the shale in one or more of the wells, pumping compressed air down the ignition well to support combustion of some of the oil shale, forcing the hot combustion gases through the oil shale to convert the solid organic matter in it to oil, and recovering the oil thus generated from other wells in the pattern.

An early investigation of this concept was made by Sinclair Oil and Gas Company, now a part of Atlantic Richfield Oil Company, which conducted experiments in 1953 and 1954 at a site near the southern edge of the Piceance Creek Basin (ref. 9). The results of these experiments indicated that communication between wells could be established through induced or natural fracture systems, that wells could be ignited successfully although high pressures were required to maintain injection rates during the heating period, and that combustion could be established and maintained in the shale bed. Additional experiments were made some years later at a depth of about 1,200 feet in the north central part of the Piceance Creek Basin. These latter tests were only partially successful, at least in part because of an inability to obtain the required surface area for heat transfer (ref. 10).

A modification of the concept shown in figure 5 was studied by Equity Oil Company of Salt Lake City (ref. 11). The modication consisted of injecting hot natural gas into the shale bed rather than having an underground combustion zone. A five-spot pattern of one injection well and four producing wells was used in an area of the Piceance Creek Basin having naturally occurring permeability and porosity due to the leaching of soluble salts. Based on results of the experiment and a mathematical model developed from them, it appeared that the technique was feasible and potentially an economic method for recovering shale oil. However, the economics are strongly influenced by the cost of natural gas and the amount required for makeup.

The only field experiment presently in progress utilizing the concept in figure 5 is being conducted by the Bureau of Mines at a site in southwestern Wyoming between the towns of Rock Springs and Green River. In this area, an oil-shale interval about 20 feet thick and yielding from 20 to 25 gallons of oil per ton is relatively shallow—50 to 400 feet deep. Over a period of several years, 10 experiments concerned with various fracturing and recovery methods have been conducted at the site (refs. 12,13). In the 11th experiment, which is presently in progress, three hydraulic fractures approximately 10 feet apart have been created. A slurried or liquid chemical explosive will be detonated in these fractures to break up the shale preparatory to an underground combustion experiment for the recovery of shale oil. Hopefully, this last phase of the experiment will start next summer or fall.

An in situ experiment where the broken shale is retorted vertically rather than horizontally is being conducted by Garrett Research and Development Company on the southwestern edge of the Piceance Creek Basin. In this technique, sufficient shale is mined from the lower part of a room to provide the desired porosity when the shale above the mined portion is fractured by explosives and collapsed into it (ref. 14). The broken shale in the room is then ignited on the top, and a combustion zone is forced down through it by supplying air to the top of the room. The hot gases ahead of the combustion zone retort the shale, and products are removed from the bottom of the room. One such room holding several thousand tons of broken shale was prepared and retorted during 1973 apparently with very satisfactory results. However, a commercial-scale application of this technique will presumably require rooms several times the size of the one completed last year and some additional development work.

A nuclear explosive, rather than partial mining, could be used to prepare a cavity filled with broken shale for in situ retorting. The concept has been discussed since the late 1950's, and a number of detailed plans for a field test have been developed—the latest in September 1973 (ref. 15). However, no field test of this technique is presently scheduled.

To furnish appropriate data for improving in situ techniques, the Bureau of Mines conducts both laboratory studies and pilot scale simulation of underground operations. The laboratory studies concern the effects of variables such as the reaction of oxygen with oil shale at subretorting temperatures, the mechanisms of formation and transport of oil out of oil-shale particles, and the effects of pressure on the retorting process. For the simulation studies, two vessels—one with a capacity of 10 tons and the other with a capacity of 150 tons—are used to study such variables as the rate of combustion front travel, gas flows through broken shale, grade of shale, and particle size distribution. The larger of the two pilot plants is shown in figure 6, which also shows in the left foreground the type of random-sized oil shale used for some experiments. This material ranges in size from sandlike particles to pieces weighing a ton or more. For experimental purposes, the retort is filled and retorting is started by igniting the shale at the top with a natural gas burner. After the burner is turned off, combustion is maintained by injecting air and recycle-gas, if used, into the top of the retort. The combustion zone travels down through the bed, retorting the oil shale ahead of it. A tank mounted on load cells is used to collect the liquid products so that a continuous record of retort output can be maintained. Gaseous products from the retort, which contain some oil and water, are passed through packed towers to remove most of the entrained materials. After passing through a blower, some of the gas stream may be recycled back into the retort, while the remainder vents through a stack equipped with a natural gas burner to oxidize combustible components. In a number of experiments run in this manner to evaluate the effects of retorting gas velocity and composition, yields up to 65 percent of Fischer assay have been obtained (ref. 16). These are thought to be promising considering the wide range in size of material being retorted and the heat losses inherent in the equipment used.

### *Oil-Shale Products*

The fuels and chemicals normally produced from petroleum can also be obtained from shale oil.

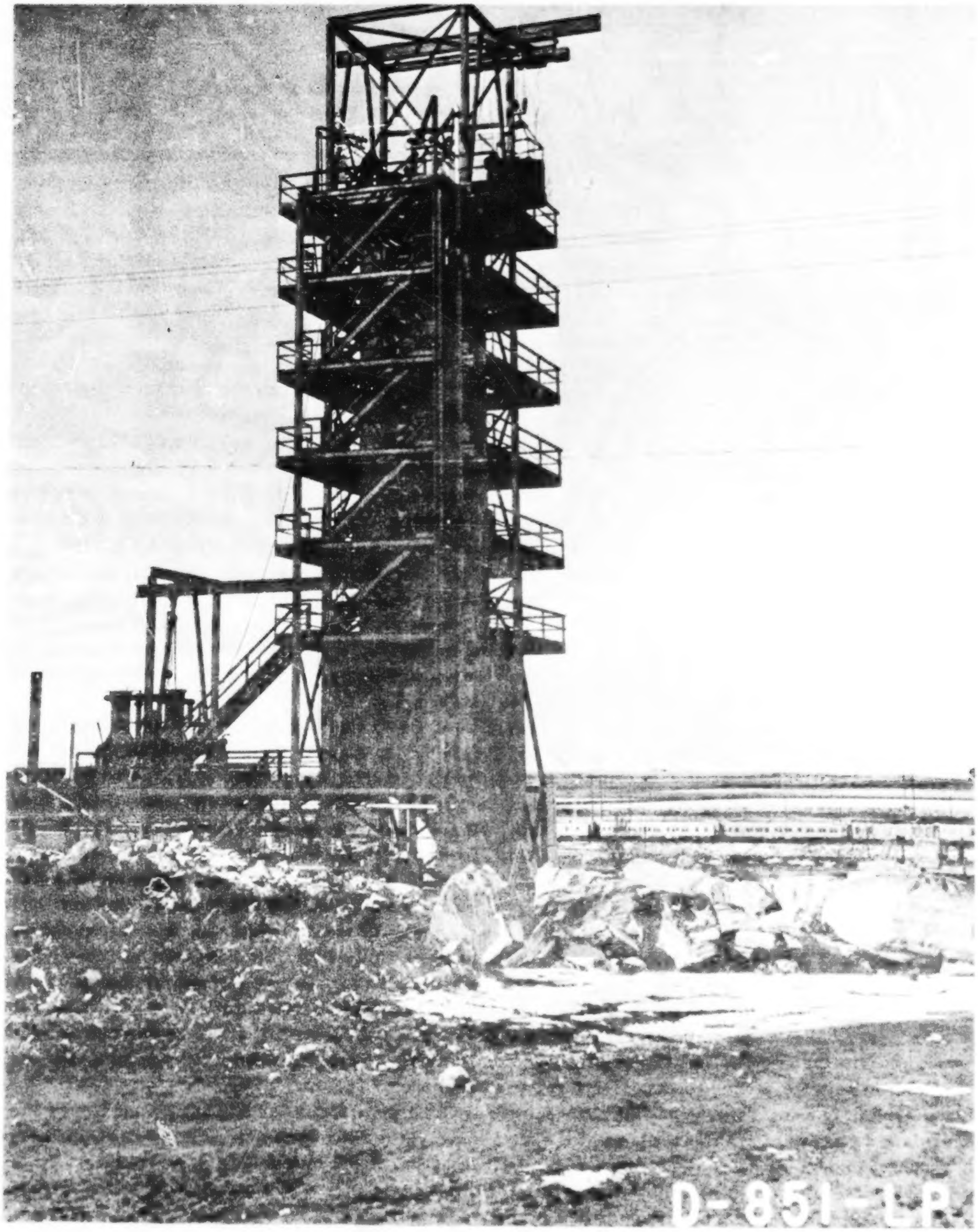

Figure 6. 150-ton retort.

Table 3. Properties of crude shale oils

| Retort | Specific gravity, 60/60° F | Pour point, °F | Viscosity, 100° F SUS | Nitrogen, wt pct | Sulfur, wt pct |
|---|---|---|---|---|---|
| Gas combustion | 0.937 | 80 | 543 | 2.16 | 0.60 |
| 10-ton | .923 | 60 | 112 | 1.57 | .79 |
| 150-ton | .909 | 60 | 98 | 1.59 | .94 |
| In situ | .885 | 40 | 78 | 1.36 | .72 |

However, an adaptation of petroleum technology based on the properties peculiar to shale oil is required. Shale oil produced by some surface retorts, such as the gas combustion retort, is usually a dark viscous material with a relatively low sulfur content but with a high pour point and a high nitrogen content (table 3). A high pour point requires that the oil receive some pretreatment before the oil is amenable to pipeline transportation. The high nitrogen content complicates the refining of the oil, so it appears that hydrotreating of the oil or some of its fractions will be required to lower the nitrogen content to acceptable levels for processing by refining methods such as catalytic cracking. In situ processing and some surface retorting systems may yield oils with low enough pour points to materially ease the problems of handling.

*Environmental Considerations*

Environmental effects directly associated with oil shale processing are expected to be from retorted or burned shales, waters that have been produced or used in processing, and gases. The present interest in in situ processing is partly because this technique would obviate the necessity of disposing of large quantities of retorted or burned shale. However, to prove in situ processing to be feasible, one must determine what effect leaching of the in place retorted shale and produced waters will have on groundwater. A start toward investigating this problem has been made by the Bureau of Mines at its Rock Springs field site where a program of well drilling and water sampling is in progress.

Although in situ processing offers some potential advantages, many portions of the Green River deposit appear to be most amenable to mining and aboveground processing as means for recovering shale oil. Hence, industry and government are both investigating problems associated with disposal of retorted or burned shale. In particular, the Colony Development Operation has done a substantial amount of work on the vegetation of retorted shale from the TOSCO II process and has shown that this revegetation can be accomplished. In another study Colorado State University in a program sponsored by industry and government, both State and Federal, established spent shale test plots at two elevations in the Piceance Creek Basin during 1973. These plots were designed to establish the requirements for germination of selected plant species and the survival rate under natural conditions.

Retorting oil shale produces water both from heating the shale and from burning the fuel when the process uses internal combustion. This water will generally be in the range of 3 to 10 gallons per ton of shale retorted. Because it has been in contact with shale oil, it contains substantial amounts of organic material in addition to inorganic ions from the minerals in the shale. Some studies of this water and suggestions for its treatment have been made (ref. 17), but additional studies will be required as more specific plans for utilization of oil shale are developed.

Gases from oil-shale processing are not expected to have a unique composition, so gas-treating methods being developed by other industries to comply with environmental requirements should be applicable to oil shale gases. However, to confirm this, postulation gases from pilot plant developments, such as those being conducted by the Bureau of Mines, should be sampled and analyzed.

In addition to the direct effects of oil-shale processing on the environment, there will be other effects from the development of an industry, particularly from the accompanying influx of people to a semiarid, sparsely populated area (ref. 18).

## RECENT DEVELOPMENTS POINTING TO COMMERCIAL UTILIZATION

Because the oil-shale deposits are about 20 percent privately owned and about 80 percent Government controlled, there is an opportunity for development on both types of land. However, since the government-controlled lands were withdrawn from leasing by President Hoover in 1930, no procedure has been developed to provide for their industrial development. In an effort to overcome this barrier, the Department of the Interior developed over the past several years a prototype leasing program which resulted in offering two leases in each of the three States—Colorado, Utah, and Wyoming—where the Green River Formation occurs. These leases, each of which is a little over 5,000 acres in size, are being offered on a monthly basis, starting with the first bid opening on January 8, 1974. The successful bidder on the first tract, which is in Colorado, was a combination of Standard Oil Company (Indiana) and Gulf Oil Corporation, with a bonus bid of a little over $210 million. The second tract, also in Colorado, went to a combination of Atlantic Richfield Company, Ashland Oil Company, Shell Oil Company, and the Oil Shale Corporation for a bonus of over $117 million. The third tract, which is in Utah, went to a combination of Phillips Petroleum Company and Sun Oil Company for a bonus bid of over $75 million. The size of these bids seems to indicate a genuine intention to develop the leases in the foreseeable future. However, this will of course depend on the future energy situation which presently is difficult to predict.

There have been two recent announcements indicating some intention to proceed with development on privately held land. The Colony Development Operation has applied for a number of the permits that will be required in order to start construction of a plant when engineering design is completed in the fall of this year. Union Oil Company of California has also announced its intention to start construction of a plant in the foreseeable future.

## SUMMARY

Green River oil shale in the States of Colorado, Utah, and Wyoming has the potential of supplying significant quantities of fuel to help fulfill the Nation's needs. Past attempts to utilize the oil shale have generally involved mining, crushing, and retorting shale aboveground. It appears that this approach will probably be applied successfully in the foreseeable future. In addition, attention is presently being given to developing in situ techniques which may have advantages, particularly from an environmental standpoint. One technique has been operated successfully in a field test and should be ready shortly for commercial-scale demonstration. The range of fuels and chemicals presently produced from petroleum can also be obtained from shale oil, if techniques appropriate to its particular composition are used. Bids recently made on leases of oil-shale land offered by the Department of the Interior and announcements made by companies holding private land both suggest that commercial development of oil shale is imminent.

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# OIL SHALE DEVELOPMENT--
# SOME ENVIRONMENTAL CONSIDERATIONS

**C. Blaine Cecil***

*Abstract*

*The primary deposits of oil shale in the United States are found in western Colorado, eastern Utah, and Southern Wyoming. The sedimentary material now referred to as "oil shale" was originally deposited over 50 million years ago in inland lakes. As a result of a complex geologic history, these sediments have been converted into sedimentary rocks rich in solid hydrocarbons. The hydrocarbon portion of the rock decomposes to oil upon heating.*

*Development of oil shale as an energy source will involve mining, crushing, and retorting. The primary products of these operations will be lowsulfur oil of commercial value and processed shale byproduct.*

*One of the environmental considerations of oil shale development consists of revegetation of processed shale; studies have shown that revegetation is feasible. A second consideration is the impact of development on water quality and quantity. Research has shown that the first commercial plant will not have a significant impact on the Colorado River system. Further data indicate that there may be as much as 400,000 acre-feet of water annually available for a mature oil shale industry in Colorado, if water rights are converted from agricultural use to industrial use. The impact of development on air quality is a third environmental consideration. Preliminary studies by the Colony Development Operation indicate that potential contaminant levels should be acceptable to both State and Federal officials.*

*As a result of technological developments in oil shale processing, coupled with environmental programs, oil from shale represents a new energy source which is available for immediate commercialization.*

The phrase "oil shale" has been used for many years to describe fine grained rocks which will yield oil when heated. "Oil shales" occur throughout the world on every continent and in at least 30 States of the United States. The world's richest reserves occur in the Eocene Age Green River Formation of Colorado, eastern Utah, and southern Wyoming as shown in figure 1.

"Oil shales" of this region are not shale, nor do they contain oil. Instead, they are more properly referred to as kerogen-bearing marlstone. These marlstones are clayey, fine-grained carbonate rocks with minor amounts of quartz and feldspars. Kerogen is the insoluble hydrocarbon protion of "oil shale" which decomposes upon heating to yield a paraffin-type "synthetic" crude oil.

Organic rich sediments which are now referred to as the Green River Formation were deposited in a lake or lakes approximately 50 million years ago. Through diageneses, these sediments were lithifield into marlstones rich in solid hydrocarbons. The organic fraction was not subjected to geologic conditions necessary for conversion to petroleum. Instead, the original organic material was transformed into the solid hydrocarbon known as kerogen.

The Green River Formation has a total areal extent of some 17,000 square miles. The U.S. Geological Survey has estimated that the total oil shale reserve of the Green River Formation in Colorado, Utah, and Wyoming is more than 600 billion barrels of oil in deposits at least 10 feet thick averaging 25 gallons or more of oil per ton of oil shale. The U.S. Department of Interior has also estimated that 80 billion barrels of this reserve are recoverable by modern mining methods. This later total is approximately twice the present domestic crude oil reserve in the United States, exclusive of Alaska, and represents approximately 75 percent as much oil as the United States has produced since the Civil War.

The Piceance Creek Basin, which will be the subject of the remainder of this paper, has an areal extent of approximately 1,250 square miles in Garfield and Rio Blanco counties in western Colorado. The Piceance Creek Basin is the richest single area of recoverable oil shale in the United States. This basin alone contains some 480 billion barrels of shale oil in reserves which are more than 10 feet thick and averaging more than 25 gallons of oil per ton of rock.

The Piceance Basin is part of the high Colorado Plateau geomorphic province with surface elevations in excess of 8,700 feet in some areas. The northern part of the basin is largely drained by Piceance Creek, which empties into the White River which in turn drains into the Green River. At the southern edge of the basin, Roan Creek and Parachute Creek drain the basin into the Colorado River. The basin is semiarid and the terrain consists of plateaus dissected by streams which have cut canyons over 2,000 feet deep

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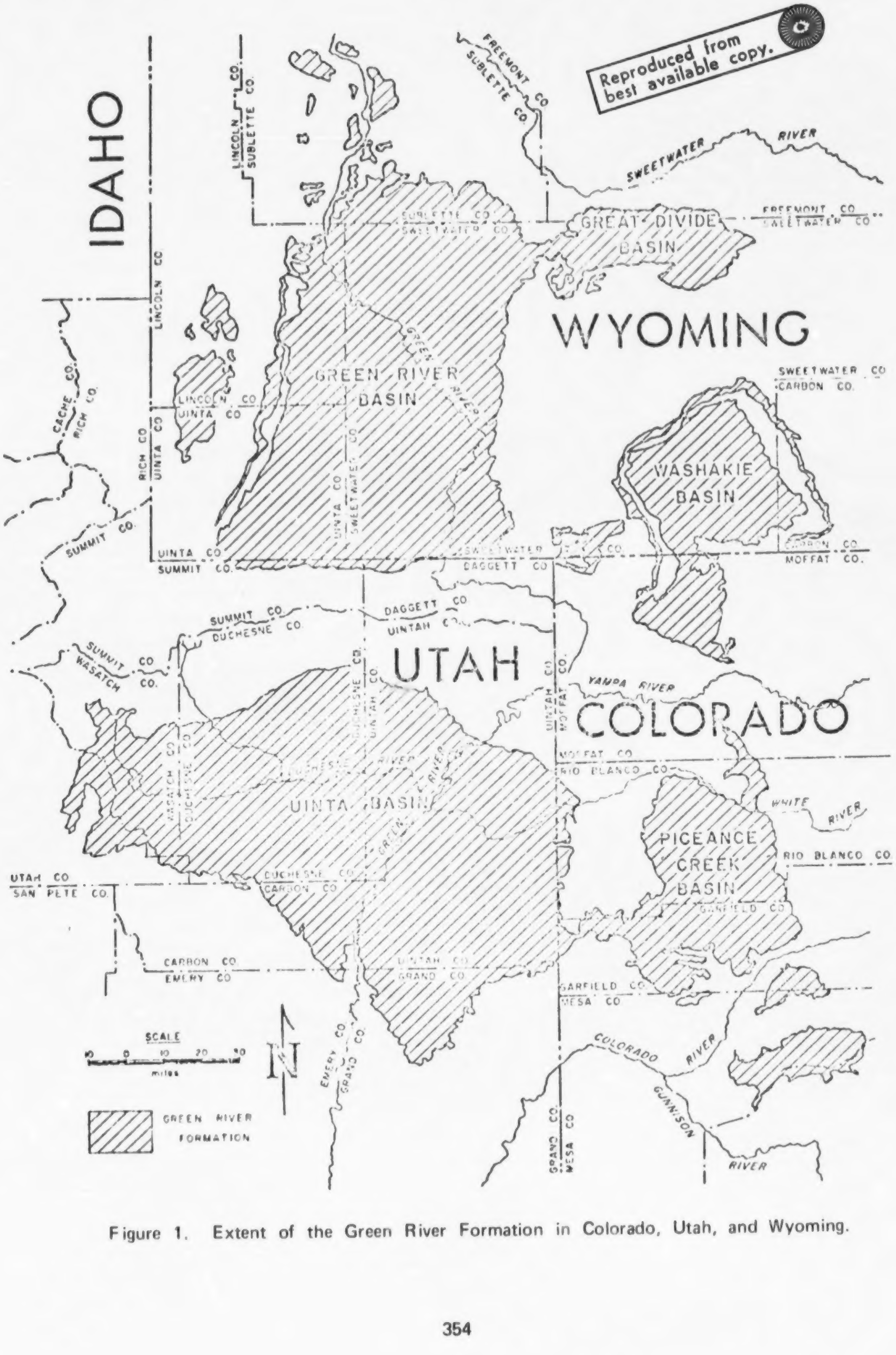
IDAHO
WYOMING
UTAH
COLORADO
GREEN RIVER BASIN
GREAT DIVIDE BASIN
WASHAKIE BASIN
UINTA BASIN
PICEANCE CREEK BASIN
SWEETWATER RIVER
YAMPA RIVER
WHITE RIVER
COLORADO RIVER
GUNNISON RIVER
GREEN RIVER
DUCHESNE RIVER
FREEMONT CO
SUBLETTE CO
LINCOLN CO
UINTA CO
SWEETWATER CO
CARBON CO
RICH CO
CACHE CO
SUMMIT CO
DAGGETT CO
UINTAH CO
DUCHESNE CO
WASATCH CO
UTAH CO
SAN PETE CO
EMERY CO
GRAND CO
MOFFAT CO
RIO BLANCO CO
GARFIELD CO
MESA CO
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N
GREEN RIVER FORMATION

Figure 1. Extent of the Green River Formation in Colorado, Utah, and Wyoming.

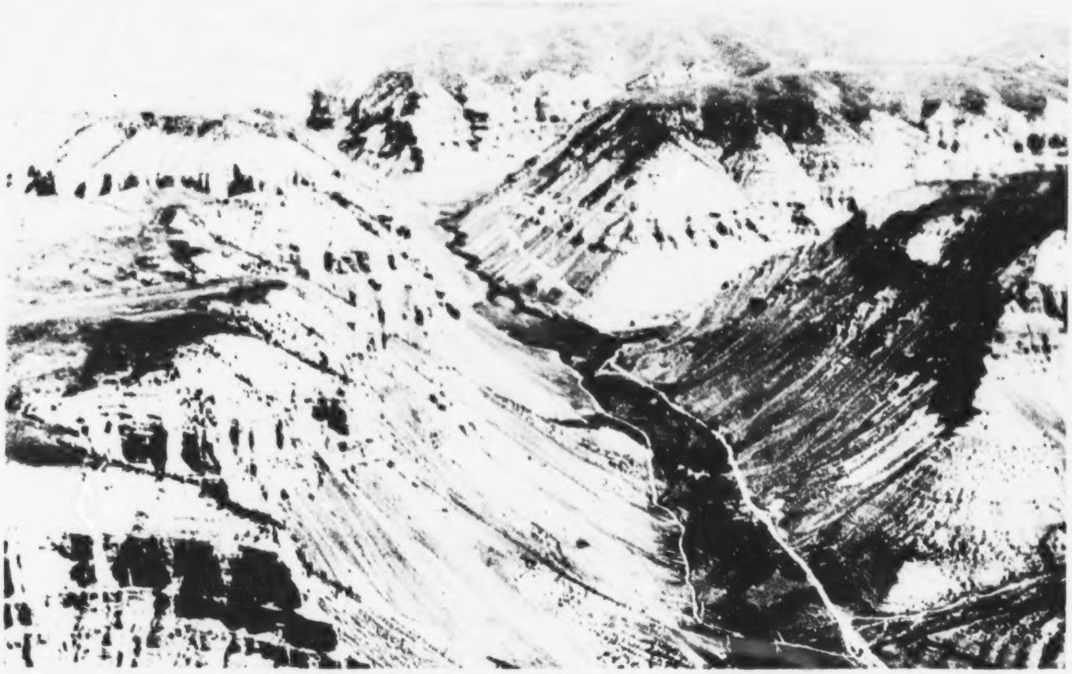

Figure 2. The upper reaches of Parachute Creek Valley, on the southern edge of the Piceance Creek Basin.

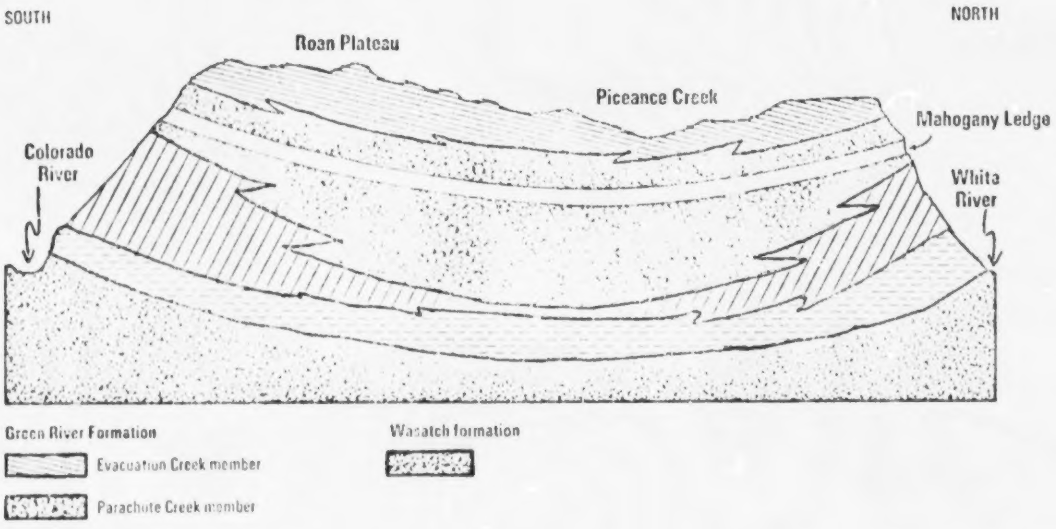
SOUTH
NORTH
Roan Plateau
Piceance Creek
Mahogany Ledge
Colorado River
White River
Green River Formation
Evacuation Creek member
Parachute Creek member
Garden Gulch member
Douglas Creek member
Wasatch formation

Figure 3. Idealized cross section of Green River Formation, Piceance Creek Basin (after Nielson).

in the southern part, as illustrated by the Parachute Creek Valley shown in figure 2.

The stratigraphy of the Piceance Basin is illustrated in figure 3. This highly simplified cross section indicates the stratigraphic position of the kerogen-rich Parachute Creek member of the Green River Formation. The Parachute Creek member contains the oil shale of greatest economic interest. As the diagram illustrates, the Parachute Creek outcrops around much of the margin of the basin and is in the subsurface in the interior of the basin. Therefore, virtually all of the oil shale zone of the Piceance Basin will be developed by underground extraction methods.

The three major components of shale oil production are mining, crushing, and retorting. A facility being planned for the Parachute Creek region by the Colony Development Operation will serve as a model for oil shale processing for the remainder of this paper. The Colony Development Operation is a joint venture consisting of four companies: The Oil Shale Corporation, Shell Oil Company, Ashland Oil, Inc., and Atlantic Richfield Company, Operator.

Conventional room-and-pillar mining will generally be utilized to extract oil shale for retorting facilities, as currently planned. The shale will be extracted from the mine, crushed, and transported by conveyor to the retorting and upgrading complex.

Retorting will be accomplished by using the TOSCO II process, the development of which began in 1956. Based on the research conducted by the Denver Research Institute of Denver, Colorado, for The Oil Shale Corporation, the first pilot plant was built near Littleton, Colorado, in 1957. In 1964, a 1,000-ton-per-day semiworks retort was constructed, and a full-scale pilot mine was started on Colony property in Middle Fork Canyon of Parachute Creek approximately 17 miles north of the town of Grand Valley, Colorado. The semiworks plant and mine were operated from 1965 through the fall of 1967. As a result of these operations, the design of a commercial oil shale complex was undertaken. Further testing of the semiworks plant was required to complete this design. The additional testing was undertaken in 1969 under the direction of Atlantic Richfield Company as Operator for the joint venture. In addition to design modifications made to the TOSCO II process, various types of pollution control equipment were evaluated. A multimillion dollar environmental program was also initiated to assess potential environmental impacts relating to air, land, water, wildlife, vegetation, and socioeconomic conditions which could result from construction and operation of an oil shale complex.

In the TOSCO II process, crushed raw shale is preheated by dilute phase fluid bed techniques. The preheated shale is then transported to a pyrolysis drum where it is mixed with hot ceramic balls. Conversion of the kerogen to hydrocarbon vapors is substantially complete when the shale reaches a temperature of approximately 900°F. Pyrolysis vapors are then condensed, fractionated, and piped to the upgrading facility for refining into final commercial products. A mixture of processed shale and ceramic balls leaves the pyrolysis drum and is conveyed to a trommel screen. The cooled ceramic balls pass over the trommel screen and are returned to a ball heater. The processed shale is cooled, moisturized, and transported to the disposal area.

The production of shale oil by retorting will generate large volumes of waste material consisting chiefly of spent shale or "processed shale." The processed shale from TOSCO's retorting process is a fine, black, powder-like material. The dark color is attributable to a small amount of residual carbon which coats the dust particles. It is powder-like because the processing temperatures are not high enough to produce the clinker-type chunks characteristic of other pyrolysis process waste materials. Because of the physical characteristics and low fertility of processed shale, special treatment of the surface embankment will be required prior to revegetation.

The environmental studies in processed shale disposal have included analyses of the permeability of the processed shale embankment, leaching characteristics of processed shale, the structural integrity of processed shale compacted to various densities, the erosion potential of processed shale, and the liquefaction potential of the embankment. The design of the embankment has been influenced substantially by the findings and recommendations set forth in these studies (ref. 1).

As an alternative to surface disposal, several studies have analyzed the potential use of processed shale for a variety of commercial purposes, including the manufacture of building blocks, concrete, road substrate, bricks, and paneling material. At the present time, none of these alternatives are economically feasible.

Revegetation of plants on the processed shale embankment is important to surface stability, reduced erosion potential, proper water balance, preservation of esthetic values, and restoration of the area to a balanced environment. For these reasons, a considerable number of revegetation studies have been conducted at the semiworks site since 1965. These programs include seven separate plot studies on processed shale which continue to provide useful information on such effects as varying slope, soil cover, mulch, and irrigation; the migration of salt through root zones; and the suitability of various local and foreign plant species for use in the ultimate vegetation process. It has been learned that regular watering during the first year is important to a rapid establishment of vegetation. Results indicate that only infrequent supplements to natural rainfall are required during the second growing season. After two seasons, the vegetation may require no further watering (ref. 1).

Successful growth of native woody plants, such as juniper, skunkbush, and four-winged saltbush in more recently established plots, indicates that these species may maintain continued growth and vigor as well as some grasses and shrubs have done in the past. The satisfactory growth of native and exotic species should not be surprising in view of the fact that processed shale is somewhat similar to the native soils derived from weathered shale. The natural soils have had many years to weather and leach; consequently, they support a wide variety of plants and vegetation types. In order to establish vegetation quickly on processed shale, the weathering and leaching process must be accelerated. This can be done by supplemental watering for a limited period of time to create a satisfactory soil medium.

### *Potential Impact on Water Quality & Quantity*

The proposed plant will use about 3.3 barrels of water per barrel of major products (fuel oil and a special quality liquefied petroleum gas) produced. Total water use will be approximately 170,000 barrels of water per day or about 11 cfs. Urban water use requirements, resulting indirectly from the population growth accompanying a commercial plant, will amount to an additional 10 to 20 percent. During normal flow of the Colorado River, this total quantity of water will be available for appropriation. During periods of low flow, unappropriated water will be obtained from other sources such as the Green Mountain Reservoir (ref. 1).

Studies of water usage have revealed that although the proposed plant is designed to be totally consumptive of water and without any effluent discharge, two potential water quality impacts may occur. Under upset or disaster conditions, discharges of effluents from the plant and mine to existing streams and aquifers could occur. In addition, slight increases in downstream Colorado River salinity concentration could result from the withdrawal of about 11 cfs for use in the plant. To the extent that this consumptive use replaces water uses which are salt loading, the net result could be a decrease in downstream salinity.

The primary use of water in the disposal operation will be to moisten processed shale, to insure maximum stability of the embankment, and to suppress dust. Colony has conducted detailed investigations of the relationship between various concentrations of water and the stability of the embankment. Preliminary studies indicate that the low permeability of compacted processed shale should minimize migration of water used to moisten the shale into subsurface aquifers. A catchment dam will be constructed below the embankment to confine runoff which may contain concentrations of salt and other dissolved solids leached from the processed shale. The confined water will be circulated back to the active disposal area for moisturizing purposes (ref. 1).

Development of oil shale in Colorado, Utah, and Wyoming may affect water quality and quantity in the Colorado River drainage basin downstream of each development site.

The historic use of water in the Colorado River Basin for various purposes including industrial development, irrigation, residential use, storage, power generation, recreation, eastern slope diversions, etc., has reduced the quantity of purer water contributed by upstream water sources and tributaries and increased the amounts of dissolved solids entering the river. As a result, salinity in the lower main stem of the river has increased steadily over the years. For example, storage of water in reservoirs throughout the basin increases evaporative loss of water by increasing the total surface area of water exposed to the atmosphere. Irrigation practices also increase the loss of water through increased evaporation and transpiration. In addition, irrigation water which is not retained within root zones or lost to deep percolation normally leaches substantial amounts of soluble salts from irrigated soil. The salt loading effect of leaching produces higher salinity in irrigation flows returned to parent streams. The totally consumptive use of water for any purpose in the upper reaches of the basin tends to increase downstream salinity levels. This is because reductions in the total amount of the relatively purer water attributable to upstream sources lead to corresponding reductions in the diluting effect of this water downstream. Water diverted for nonconsumptive industrial, municipal, and residential purposes is normally returned to the parent streams with a higher salinity concentration than at the original point of diversion. The concentrating effect of the evaporative losses and salt loading related to all of these uses currently threatens to diminish the range of uses to which lower basin water can be applied.

Despite the fact that the general sources of increased salinity have been identified, cause and effect relationships between specific upstream uses of water and increased downstream dissolved solid concentrations are difficult to define precisely. Because of the mingling of water from separate sources in basin reservoirs and the release of water stored in these reservoirs for varying lengths of time, the specific impact of any single upstream facility or use is difficult to isolate. According to the Department of State (ref. 2), the resulting storage "damping" effects of upstream actions on downstream water quality may be spread over several years. Therefore, it may be impossible to identify the specific effect of any individual upstream use in any quantifiable way downstream.

The potential deterioration of water quality in the lower main stream of the Colorado River has become the subject of continuing negotiations between the United States and Mexico. In order to insure compliance with the various agreements between these countries on salinity, the Department of State has considered various alternative for controlling salinity concentrations at the United States-Mexican border. The environmental impacts of each of these alternatives have also been compared and evaluated (ref. 2). Subject to the passage of enabling Federal legislation, the Department of the Interior has elected to construct desalting facilities in the lower portion of the basin as the best solution to the problem at this time. Various international, Federal, multistate, and State advisory groups are searching for independent solutions. The following salinity policy statement, issued by the Seventh Session of the Enforcement Conference on Pollution of the Colorado River, has received favorable comment from various Federal and State agencies:

> "A salinity policy be adopted for the Colorado River System that would have as its objective the maintenance of salinity concentrations at or below levels presently found in the lower main stem. In implementing the salinity policy objective for the Colorado River system, the salinity problems must be treated as a basin-wide problem that needs to be solved to maintain Lower Basin water salinity at a below present levels while the Upper Basin continues to develop its compact-apportioned waters." (ref. 3)

As yet, neither the Federal Government nor the State of Colorado has established maximum values or standards for salinity concentrations in natural water bodies or for effluents from industrial, municipal, or agricultural sources. Because the Environmental Protection Agency presently has the authority to prescribe standards for most types of discharge, it is expected that regulation of effluents (including irrigation return flows) will precede regulations applicable to the indirect effects of totally consumptive diversions.

A comparison of the volume of Colony's expected diversion for the proposed plant north of Grand Valley, Colorado, with the volume of other existing or proposed uses, is helpful to the overall evaluation of the significance of the impact of the Colony project. Colony presently intends to consume less than 9,000 acre-feet of water per year at the proposed plant and mine. According to the Denver Water Board, the City and County of Denver plan to divert up to 183,000 acre-feet of water per year from the Colorado River Basin by 1980 and up to 338,000 acre-feet per year by the year 2000. Evaporative loss of water per year from Lake Mead exceeded 700,000 acre-feet during the years 1970, 1971, and 1972. Considering the magnitude of evaporative losses from other basin reservoirs, such as Lake Powell and at Imperial Dam, the salinity impact of the Colony project will be negligible (ref. 1).

Based on the previously quoted recommendations of the Enforcement Conference on Pollution of the Colorado River, Skogerboe (ref. 3) has proposed a policy of nondeterioration of present salinity levels by maintaining a net salt balance at the point of diversion in Grand Valley equivalent to the present mean annual salinity concentration at Hoover Dam. Inherent in this proposal is the assumption that the salinity level at Hoover Dam should be the controlling value; depending upon the goal of any particular user's policy, other control values could be selected. The possibilities include the mean annual concentration at Imperial Dam, at other downstream locations in the watershed, or the value at any point in the basin which constitutes the limiting value for any specific existing use.

Nondeterioration is only one of a number of alternate methods for controlling downstream salinity. These methods include the construction of desalting facilities, more efficient use of water in irrigation operations and substitution of water obtained from sources outside the basin (refs. 3,4). As indicated previously, the Federal Government has selected the alternative of desalting plants. This alternative has the advantage of preserving the full range of existing uses of water in the upper portion of the basin.

In view of the complexity of the issue of salinity control, the likelihood of future regulation, and the number of alternate control methods, it is inadvisable for an individual water user to make significant expenditures to implement any single alternative at this time. As a result, Colony intends to continue monitoring the development of salinity controls and to evaluate further the need for mitigating the slight impacts of the proposed plant before commiting itself to any particular solution.

Substantial quantities of ground water are believed to exist under the two Federal test lease tracts of the Piceance Basin. It may be possible to use the water produced by mine dewatering in plant operations associated with the Federal lease tracts. If this is possible, consumptive use on lease tracts from surface drainages may be substantially reduced.

The question concerning water availability for various uses in the Colorado River Basin is ongoing. This question has also been asked regarding the new oil shale industry. Sparks (ref. 4) concluded that 250,000 acre-feet of water can be made available to support a mature oil shale industry in Colorado. This amount could perhaps be increased to as much as 400,000 acre-feet with a corresponding loss in agricultural production, because most of the available water supplies in Colorado are devoted to agriculture according to Sparks (ref. 4).

The possible environmental impact of an oil shale processing facility on air quality will consist of minor effects such as slightly higher local surface temperatures, increased local humidity, minor interference with existing diurnal wind patterns, and a slight reduction in local solar radiation. In addition, there may be increases in ambient levels of sulfur oxides, nitrogen oxides, particulates, hydrocarbons, and carbon monoxides resulting from plant stack emissions as well as the generation of fugitive dust. However, the Colony Development Operation believes that air contaminants which may be released from a commercial plant will be acceptable to both Federal and State authorities. Such conclusions are based on studies which were carried out at Colony's site on Parachute Creek (ref. 1).

Due to limited time, there are various environmental considerations which have not been discussed in detail in this paper. However, past studies have been numerous and comprehensive. With the leasing of Federal lands in the Piceance and Unita Basins, numerous new detailed studies are being undertaken by the various companies involved in the leasing program. The past and ongoing studies have and will continue to define potential problems as well as plausible solutions. The result of all the environmental research will allow maximum development with minimum impact. As a result of technological developments coupled with environmental considerations, oil shale now represents a viable alternative energy source which is available for immediate commercialization. The low-sulfur fuel oil which will likely be produced by the initial commercial plant (expected to under construction in Colorado within a year) has obvious environmental benefits in the regions of the country where it will be consumed. The companies directly involved with oil shale development believe that oil from shale is a partial solution to the current energy crisis.

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# OVERVIEW OF R & D NEEDS ON ENVIRONMENTAL ASPECTS OF COAL-CONVERSION PROCESSES

**A. A. Jonke and W. Podolski***

*Abstract*

*This paper gives an overview of the environmental factors related to all of the various types of coal-conversion processes, and it considers the R & D needs for alleviating environmental concerns. The research requirements for avoiding serious environmental abuse are vast and range from improvements in coal mining methods to the problems of disposing of waste products and heat. The conversion of coal to liquid or gaseous fuels does not automatically assure an improved environment. Pollutants, such as ash and sulfur, must be removed from the coal-derived fluids to levels even lower than those corresponding to the emissions from conventional coal-burning power plants. Unless more efficient power cycles are developed, the overall thermal effects of coal conversion may be greater than those for conventional power plants. The quantification of the environmental problems for each coal-conversion process is needed, and the commonalities and differences of the various processes must be weighed. A systems analysis study will be needed to provide the basis for intelligent planning for the utilization of coal energy with minimum adverse environmental, health, and safety effects.*

## INTRODUCTION

The expected emphasis on and drive toward national energy self-sufficiency implies a very real possibility for large scale environmental degradation. There is great concern, on the one hand, that unabated growth in energy production and utilization will cause irreparable harm to the environment and, on the other hand, that measures to protect the environment may further exacerbate the current supply problems faced by the energy industry. The environmental costs of vastly increasing domestic energy supplies are hard to calculate, but they are potentially severe.

It is now generally recognized that the Nation must use coal over the next few decades at rates that are double or treble our current levels. Our ability to actually achieve such an expansion of production and utilization is in serious doubt, mostly due to uncertainties about environmental impacts. The research requirements for avoiding serious environmental abuse are vast and range from needed improvements in coal-mining methods to the problems of disposing of waste products and heat. We should first recognize, however, that many of the environmental problems may not yet be known.

The major emphasis in coal conversion R & D, thus far, has been on the solution of technical problems; as a consequence, there are large gaps in our knowledge of the fundamental processes that are involved in coal extraction, conversion, and utilization. Most of the schemes for producing clean forms of fuel from coal that have progressed beyond the pilot-plant stage are based on upgraded technology of the 1930's, and many of the potentially attractive schemes for conversion of coal to alternative fuels are still at a bench scale. In many cases, we are building pilot plants and designing demonstration plants on the basis of empirical art. This can lead to unforeseen environmental consequences when the processes are scaled up.

Pilot plants are generally operated over a limited range of variables in order to optimize operating conditions and to provide economic data for subsequent commercial-scale operation. However, the information needed for determining all of the necessary environmental ramifications may not be obtainable from pilot-plant programs on a timely basis. Hence, one important need related to environmental impact of coal conversion will be a carefully thought-out fundamental program, including bench-scale studies, aimed at identifying and quantifying all of the environmental problems that require solution.

Although it now appears that substantial Federal and private funding will become available to conduct needed R & D, trained manpower with experience in this field is in short supply. Consequently, there will be a need to utilize scientists and engineers from related fields. A cooperative effort that employs Federal laboratories, private industry, and universities in the most effective manner will be highly desirable.

This paper is intended to present an overview of R & D requirements for coal conversion processes. But

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to avoid repetition and conflict with other presentations in this symposium, we will concentrate on selected areas and overall systems requirements that are common to all coal conversion and utilization schemes. Specific R & D needs associated with various fossil-fuel conversion schemes are covered by several very competent speakers in this session.

## THE IMPORTANCE OF ADVANCED POWER CYCLES

The conversion of coal to liquid or gaseous fuels does not automatically assure an improved environment. Pollutants, such as ash, sulfur, and nitrogen compounds, must be removed from the coal-derived fluids to levels even lower than those corresponding to the emissions from conventional coal-burning power plants. A substantial R & D effort will be needed to accomplish the cleanup of these fuels in an efficient and economical manner. Consider, also, the matter of waste heat. The realization that the Nation's sources of water for cooling may not be great enough to fill energy requirements in the decades ahead makes the waste heat problem one of increasing concern. A modern fossil-fuel power plant operating at 41 percent efficiency gives off 1.4 kW of waste heat for each kW of electricity produced. If we convert the coal to a fluid fuel at a thermal efficiency of perhaps 85 percent the amount of waste heat emitted jumps to nearly two units per unit of electricity. Fuel requirements increase in like manner.

Hence, the use of coal conversion schemes in conjunction with conventional steam-power cycles does not appear to be one of the most attractive options, except perhaps on an interim basis. Instead, major incentives exist for a large R & D effort to develop more efficient power cycles that take full advantage of the potential of clean fuels.

Advanced power cycles, if successfully developed, would conserve our fuel resources and decrease air, water, and thermal pollution. But what types of advanced power cycles should we develop? A number of systems have been under study and currently are in various stages of development. These include: combined gas/steam cycles, potassium topping cycles, Rankine bottoming cycles, closed cycle helium turbine systems, plasma MHD, liquid metal MHD, supercritical $CO_2$ cycles, and others. Although their advantages and disadvantages have been discussed in the literature, it is not possible, from the information available, to assess the relative merits of the various concepts. A number of studies have been made that allege to compare the various conversion cycles. However, conditions are generally picked to emphasize positive results of a specific system that is being promoted, whereas, minor changes in the ground rules of such studies could change the conclusions reached.

The first important need, therefore, is for a comprehensive and careful systems analysis of all the important concepts and fuel alternatives. The objective of such a study would be to provide guidance in evaluating the advanced conversion options, establish program priorities, and develop funding requirements. This study should be designed to evaluate the competitive positions of the various concepts based on a comprehensive, consistent set of evaluation factors. Such evaluation factors might include the probable performance of the technology, the utility of the technology, the R & D requirements, the resources required to achieve utilization, and the impacts of the technology.

On the basis of a recent advertisement in the *Commerce Business Daily*, it appears that an evaluation study of advanced energy-conversion systems is about to be launched under management of the NASA Lewis Research Center. After priorities are established, R & D to develop the improved cycles should be carried forward expeditiously. There is a real expectation that cycle efficiencies of 50 percent or more can be achieved.

We need also to explore the practicality of new bottoming cycles, using working fluids like ammonia, isobutane, nitrogen tetroxide, etc. This technique might further reduce fuel requirements, waste heat emissions, and other environmental effects. Although such bottoming cycles have been considered in the past, the changing energy picture warrants a new look at this concept.

## SOME COAL EXTRACTION CONSIDERATIONS

We turn now to another serious problem area that is common to all processes that utilize coal--namely, extracting it from the earth. Deep coal mining, as presently practiced, is a dirty and hazardous business. Strip mining can be a disaster to the landscape. As the scale of mining activities increases, the problem becomes ofincreasing national concern.

Consider, for example, a hypothetical situation in which all the Nation's energy were supplied by strip-mined coal. The demand would be about 5 million tons per day. For example, if an 18-inch thick seam (a very worthwhile deposit) were processed, about 3 square miles per day would be overturned to supply the demand, and a comparable amount might be destroyed by spoil banks, access road, etc., under present practices (ref. 1).

The consequences of surface mining practices are well known. In the eastern part of the country, exposed seams and spoil banks cause acid drainage that ruins waterways and spoils fishing. The fragile land may be softened by rainfall thus causing whole disturbed hillsides to slide, scarring large areas. In the West, alkaline soils and insufficient rainfall and nutrients to insure vegetation are the chief problem. The very thin topsoils in the West cannot easily be segregated for reuse, or if that problem is solved, they cannot be held in place.

Cost factors, to a large extent, will determine the future of strip mining because of stringent reclamation requirements that must be imposed on these operations. New concepts need to be developed in order for the surface mining industry to meet its environmental obligations. Research and development needs can be grouped into two categories: first, mining methods and equipment development, and second, improved land reclamation measures.

In the area of improved mining techniques, several methods (ref. 2) could have a positive environmental effect. One of these is the use of highly mobile draglines in conjunction with bulldozers in order to level the tops of spoil banks and to decrease the time lag between mining and reclamation. A second method may be to use a belt transfer system to move the spoil around the working areas. Each of these methods and many others not mentioned will need specialized pieces of equipment, often on a site-specific basis. A large scale development effort is needed here.

Special effort is needed to develop the means to mine the very thick western seams. These seams are too thick to provide sufficient roof support for underground mining, and they stress the limits of conventional surface-mining equipment. Combinations of surface and underground mining and improved open-pit mining operations should be investigated.

Extensive development needs exist in the area of reclamation and reuse of surface-mined lands. In a way, this may be fortunate because large increases in coal production are likely to be from surface mines, and the environmental effects from these mining methods are reasonably well known and can be controlled, albeit at a high cost. Land reclamation is also likely to be site specific, thus adding to the cost. For the western States, research needs to be done on the possibility of providing artificial soils with various agents such as enzymes and bacteria to enable plants to grow. Fast growing plants capable of growing in highly alkaline soils need to be developed. In the East, it will be necessary to develop low-cost means of neutralizing acid soil. Some small scale work is being done (ref. 3) on the use of sewage sludge in conjunction with agricultural lime to assess the ability to reclaim surface-mined land in Illinois. Since this effort has shown promise, an expanded effort is needed to develop new techniques for revegetating spoil areas.

Since the development of environmentally sound surface mining techniques will be very costly, it is essential that models and simulations of strip-mining operations be developed. Since equipment is mostly site specific, a model would enable variables in surface mining to be optimized as they relate to a given region. By simulating surface mining operations, land reclamation costs can be included, and development costs can be minimized. Limited field studies will be necessary to provide quantitative data for use in the simulations. These studies should all be part of an overall evaluation of the alternative means of obtaining energy coupled with intelligent land and resource use planning.

There are perhaps as many areas for potential research related to underground mining as there are for surface mining. Details on many of these areas have been discussed in the report of the Carnegie-Mellon workshop on Advanced Coal Technology (ref. 4). While the environmental effects of surface mining are reasonably well understood, as mentioned above, the long-term effects from surface subsidence resulting from underground mining are neither well understood nor readily predictable. It is necessary to investigate the causes and factors affecting surface subsidence in order to be better able to predict its occurrence and effects. Development of techniques for flushing or backfilling is also necessary. The benefits that would ensue are the general structure of the land would be preserved, and communication between surface and subsurface water would be cut.

A major investigation is needed that would be directed at preventing the formation of acid mine water to avoid the need for the remedial type treatment now used. Other areas needing research work include development of techniques for recovering coal fines to improve coal economics and reduce air pollution, improving sulfur removal at the coal preparation plant, perhaps by chemical means such as leaching, and finally finding means for high volume utilization of solid mine wastes.

## OVERALL SYSTEMS NEEDS

Systems planning for the conversion of coal into alternative fuels presupposes that a decision has been made to utilize the Nation's coal resources in a particular manner. In fact, however, a need exists to develop an overall plan on both a national and regional basis for the utilization of our fossil fuel resources. Regional plans could then provide answers concerning the best use of Colorado River water and the ultimate use for the reclaimed land. Long-term plans could be made, for instance, to return western lands to grazing pasture or recreational purposes or even to eliminate certain coal lands from production. Intelligent, long-range land and resource use plans need to be formulated, at least on a regional level, and it is in the framework of regional planning that waste heat disposal and water utilization are best placed. Here, research in setting up integrated models or projects is desirable, and a high priority should be given to such efforts.

As an illustration of the systems needs, means of predicting the effects of waste heat disposal is obviously needed, for, in the face of this uncertainty, very strict water temperature standards are being adopted at an enormous cost. Thus, high priority items are (ref. 5) research to predict water temperatures in a specific zone that result from various discharges and the biological effects associated with such changes. Some intelligent criterion must also be developed to weigh the cost of possible damages against the cost of preventing any damages.

Research should also be encouraged to find beneficial uses of waste heat. Possible applications, which need more investigation, are the use of heat to enhance fish culture or to use it in agricultural applications.

The availability of water will undoubtedly limit the growth of a coal conversion industry in the western States. The problem can only be addressed on a regional basis. Questions concerning what happens to the water supply if large quantities of coal are removed from the ground must still be answered. Much development is needed to minimize the consumptive use of water in converting coal to alternative fuels. Dual purpose utilization of water may have to be instituted. For example, water destined for irrigation might first be used for process cooling water. It is clear that conventional economic factors and market action will not be enough to bring about the necessary improvements in efficiency.

Technical, economic, and environmental impact assessments covering the entire field of coal-based energy technologies are urgently needed. The optimum utilization of coal energy cannot be accomplished without examining the total energy picture and identifying the relative impacts that coal will have.

## CONCLUSION

We have briefly touched on some areas relating to coal conversion research and development needs, and we have been by no means exhaustive, but rather selective, in their treatment. It is very probable, as we have pointed out, that the size of a coal-conversion industry will be limited not so much by environmental objections to the processes themselves, but by limitations placed on related areas such as mining capacity in the face of stringent land reclamation requirements. The chief emphasis of our presentation is that research relating to overall systems concepts should be given the highest priority, otherwise development of a coal conversion industry will be on a haphazard basis, and it will experience many delays while contributing little to increased utilization of our fossil fuels. The delays currently being experienced by the nuclear power industry, as a result of environmental concerns, should be kept in mind.

Finally, many of the problems that we have discussed relating to coal conversion also apply to the increased utilization of our oil-bearing shale formations. Land use and water availability problems are even more severe for shale utilization, so that overall systems planning is exceedingly important.

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# ERRATA SHEET

for the

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NOTE: Please mark your proceedings volume with all changes indicated on the reverse side of this sheet first; then proceed to the item given below.

| Page | Table No. | Changes |
|---|---|---|
| 73 | 4 | • Paste the replacement table below over the table 4 printed in the proceedings volume: |

Table 4. Elements of Current U.S. Regulatory Policy for Air and Water Pollutants

| Elements of Air Pollution Control Policy | Elements of Water Pollution Control Policy |
|---|---|
| AMBIENT AIR QUALITY STANDARDS | RECEIVING WATER QUALITY STANDARDS |
| State and Local Standards | State and Local Standards |
| Nat'l Primary Standards | |
| Nat'l Secondary Standards | |
| Nondegradation Standards | |
| -- | U.S. PUBLIC HEALTH SERVICE DRINKING WATER STANDARDS |
| THRESHOLD LIMIT VALUES | -- |
| EMISSION STANDARDS | EFFLUENT LIMITATIONS |
| State and Local Standards | State and Local Standards |
| New Source Performance Standards | New Source Performance Standards |
| | Best Practicable Technology Currently Available |
| | Best Available Technology Economically Achievable |
| | Pretreatment Standards |
| | Zero Discharge |
| Hazardous Pollutants | Toxic Substances |

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## ERRATA SHEET/March 14, 1975 – for EPA-650/2-74-118

| Page | Table No. | Changes |
|---|---|---|
| 75 | 6 | • On the line marked "Kentucky", in second column headed by "Pollutant", spelling should be "Fluorides". |
| | | • In third column heading, change "ug/m$^3$" to "$\mu$g/m$^3$". |
| | | • In the grouping marked "Pennsylvania", in the second column headed by "Pollutant", change the line marked "Sulfates ($H_2SO_4$)" to read "Sulfates (as $H_2SO_4$)" (add the word "as"). |
| | | • At end of first footnote, add: "(Ref. 34)". |
| 75 | 7 | • In second and third columns, change "ug/m$^3$" to "$\mu$g/m$^3$" (13 times). |
| 77 | 8 | • On the line marked "Ohio", in third column, change "0.1 TLm-48 hour" to "0.1 $TL_m$-48 hour" (make "m" a subscript). |
| | | • On the line marked "West Virginia", in third column, change "0.1 $Tl_m$-96 hour" to 0.1 $TL_m$-96 hour" (make "l" a capital L). |
| | | • In the space below the acronym "ORSANCO", add the definition of ORSANCO: "(Ohio River Sanitary Commission)". |
| | | • Add an asterisk "*" to end of table title. |
| | | • Add footnote under table: "*Refs. 16, 32". |
| 78 | 9 | • Under "Water Pollutants" section, on eighth line headed "phenols", add an "x" in the column headed by "Iron Making". |
| 79 | 10 | • Title should read: "Proposed New Source Performance Standards for Petroleum Refining***" (add the word "Proposed" and add three asterisks "***" after title). |
| | | • Transpose the entire second column, *including its heading and data*, to the right of the third column so that left to right the columns will be "One Day Max* Range" and "30 Day Max** Range". |
| | | • Add third footnote: "***Ref. 20". |
| 79 | 11 | • Title should read: "Proposed New Source Performance Standards for Byproduct Coke Making***" (add the word "Proposed", and add three asterisks "***" after title). |
| | | • Add third footnote: "***Ref. 21". |
| 80 | 13 | • In the second column heading, following "WATER", add a double asterisk "**". |
| | | • Delete the footnote, "*For specific sources only", and replace it with two footnotes:<br>"*Ref. 33<br>"**Ref. 29". |
| 81 | 14 | • On first line marked "Illinois", in the sixth column headed with "$SO_2$ (ppm)", move "19.5 $P^{0.67}$ lb/hr (P = tph wgt rate)" down to the space below, on line marked "Indiana"; be sure to add a parenthesis "(" in front of the "P", and change "wst" to "wgt". |
| | | • In its place, on the line marked "Illinois", add "2000". |
| | | • On last line marked "West Virginia", in the fifth column, change "21.2-50.0" to "21.2*" (i.e., delete "-50.0", and add an asterisk "*"). |
| | | • Add double asterisks "**" to end of table title. |
| | | • Add a second footnote: "**Ref. 34". |
| 81 | 15 | • In table title, delete the word "Proposed" and substitute the words: "State of". |
| | | • In the second column, on the second line, change "0.15 lb/MMBtu" to "0.16 lb/MMBtu" (change "0.15" to "0.16"). |
| | | • In the third column, on the first line, change "0.03 lb/MMBtu" to "0.03 gr/scf". |
| | | • In the first column, on the fourth line, delete "Non-methane Hydrocarbons" and replace it with "Hydrogen Sulfide". |
| | | • In the third column, on the fourth line, change "Nil" to "10 ppm". |
| | | • In the first column, on the fifth line, change "Sulfur (Vapor)" to "Total Sulfur". |
| | | • In the third column, on the fifth line, change "0.04 lb/MMBtu**" to "0.008 lb/MMBtu". |
| | | • Delete the second footnote: "**Becomes 0.008 December 31, 1978". |
| 88 | 17 | • On the bottom line marked "Total USA", in the fifth column, change "4255" to "14,255". |
| | | • Remove the word "the" from the table title. |